

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
BEFORE THE HONORABLE BOARD OF PATENT APPEALS AND
INTERFERENCES

In re the Application of

Johannes BOS

Application No.: 10/580,018

Examiner: G. LISTVOYB

Filed: May 19, 2006

Docket No.: 127898

For: PROCESS FOR MAKING DAPBI-CONTAINING ARAMID CRUMBS

BRIEF ON APPEAL

Appeal from Group 1796
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I. REAL PARTY IN INTEREST

The real party in interest for this appeal and the present application is Teijin Aramid B.V., by way of (1) a Change of Name recorded in the U.S. Patent and Trademark Office at Reel 020600, Frame 0784 and (2) an Assignment recorded in the U.S. Patent and Trademark Office at Reel 017934, Frame 0461.

II. STATEMENT OF RELATED CASES

There are no prior or pending appeals, interferences or judicial proceedings, known to any inventor, any attorney or agent who prepared or prosecuted this application or any other person who was substantively involved in the preparation or prosecution of this application, that may be related to, or that will directly affect or be directly affected by or have a bearing upon, the Board's decision in the pending appeal.

III. JURISDICTIONAL STATEMENT

The Board has jurisdiction under 35 U.S.C. §134(a). The Examiner mailed a Final Rejection on June 15, 2009, setting a three-month shortened statutory period for response. The time for responding to the Final Rejection expired on September 15, 2009. Rule 134. A Notice of Appeal was filed on September 15, 2009. The time for filing an Appeal Brief expires the later of two months from the filing of the Notice of Appeal, or one month from the mailing date of the Notice of Panel Decision if a Pre-Appeal Brief Request for Review is sought. Bd.R. 41.37(c) and Official Gazette Notice, July 12, 2005. No Pre-Appeal Brief Request for Review was sought. The extendible period for filing the Appeal Brief therefore expires November 16, 2009 (November 15 being a Sunday). This appeal brief is being timely filed on November 16, 2009.

IV. TABLE OF CONTENTS

	<u>Page</u>
I. Real Party in Interest.....	2
II. Statement of Related Cases.....	3
III. Jurisdictional Statement.....	4
IV. Table of Contents.....	5
V. Table of Authorities	7
VI. Status of Amendments	8
VII. Grounds of Rejection to be Reviewed.....	9
VIII. Statement of Facts.....	10
IX. Argument.....	24
A. Introduction	24
1. Overview Of Rejections.....	24
2. Overview Of The Claimed Subject Matter	24
B. Claims 1-5 And 10 Would Not Have Been Obvious Over Vollbracht in View of Chernykh And In Further View Of Jung.....	25
1. Vollbracht, Chernykh And Jung Do Not Describe The Limitations Recited In The Independent Claims.....	25
2. Appellant Has Demonstrated That Obtaining A Crumb Directly During The Copolymerization of PPD, DAPBI And TDC Monomers Is Not A Matter Of Routine Experimentation	26
3. The Cited References Provided One Of Ordinary Skill In The Art With No Reasonable Expectation Of Success In Forming A DAPBI PPTA Aramid Crumb	34
4. The Examiner's Allegation That Chernykh Describes The Equivalence Of LiCl And CaCl ₂ Would Be Applicable To Crumb Formation Is Incorrect.....	35

5.	The CaCl ₂ /NMP Salt/Solvent Combination Described In Table 2 Of Chernykh Is Directed To A Different Chemical Compound Than Recited In Claims 1 And 10	36
6.	The Examiner's Allegation That The Evidence Presented In The Rule 132 Declaration Regarding The Alleged Equivalence of Lithium Chloride And Calcium Chloride Is Not Commensurate In Scope With The Independent Claims Is Incorrect	36
7.	Conclusion.....	37
C.	Claims 2-5 Would Not Have Been Obvious Over Chernykh Or Jung in View of Vollbracht Or Encyclopedia	37
X.	APPENDIX A - Claims Section.....	40
XI.	APPENDIX B - Claim Support and Drawing Analysis Section	44
XII.	APPENDIX C - Means or Step Plus Function Analysis Section.....	47
XIII.	APPENDIX D - Evidence Section.....	48
XIV.	APPENDIX E - Related Cases Appendix	106

V. TABLE OF AUTHORITIES

<u>Cases</u>	<u>Page</u>
<i>CFMT v. Yieldup Intern. Corp.</i> , 349 F.3d 1333 (Fed. Cir. 2003)	22
<i>In re Corkill</i> , 771 F.2d 1496 (Fed. Cir. 1985)	36
<i>In re Fay</i> , 347, F.2d 597 (C.C.P.A 1965)	23
<i>In re May</i> , 574 F.2d 1082 (CCPA 1978)	30
<i>In re Ochiai</i> , 71 F.3d 1565 (Fed. Cir. 1995)	22
<i>In re Wada and Murphy</i> , Appeal 2007-3733 (2007)	22
<i>Pfizer, Inc. v. Apotex, Inc.</i> , 82 USPQ 2d 1321 (Fed. Cir. 2007)	23, 36
<i>Sanofi Synthelabo Inc., v. Apotex, Inc.</i> , 89 USPQ2d 1370 (Fed. Cir. 2008)	24
<u>Statutes</u>	
35 U.S.C. §103(a)	9

VI. STATUS OF AMENDMENTS

No Amendment After Final Rejection has been filed.

VII. GROUND OF REJECTION TO BE REVIEWED

The following grounds of rejection are presented for review:

1) Claims 1-5 and 10 are rejected as having been obvious under 35 U.S.C. §103(a) over U.S. Patent No. 4,308,374 ("Vollbracht") in view of RU 2017866 ("Chernykh") and in further view of U.S. Patent No. 5,646,234 ("Jung"); and

2) Claims 2-5 are rejected as having been obvious under 35 U.S.C. §103(a) over Chernykh or Jung in view of Vollbracht or Encyclopedia of Pol. Sci. and Tech, Polyamides, vol. 3, p. 565-567 ("Encyclopedia").

VIII. STATEMENT OF FACTS

1. The present claims relate to a method for obtaining an aromatic polyamide ("aramid") composition and an aramid composition comprised of para-phenylene terephthalamide ("PPTA") and 2-(p-phenylene)benzimidazole terephthalamide units by copolymerizing para-phenylenediamine ("PPD"), 5(6)-amino-2-(p-aminophenyl)benzimidazole ("DAPBI"), and terephthaloyl dichloride ("TDC") in a mixture of N-methyl pyrrolidone ("NMP") and calcium chloride ("CaCl₂") (Claims 1 and 10; Specification ("Spec"), page 1, lines 1-6).
2. The aramid composition is in the form of a crumb or crumb-like material formed directly during the copolymerization of the PPD, TDC and DAPBI monomers (Claim 1; Claim 2; Spec, page 1, lines 30-31; page 2, lines 13-17; and page 3, lines 20-37).
3. Crumbs are desirable because crumbs can be easily formed into a spin dope, which can be further processed into fibers and films (Spec, page 3, lines 17-19).
4. The term "crumb" or "crumb-like" means that the polymer mixture contains breakable clumps or particles that (1) are non-sticky and (2) have a mean particle size greater than 100 μm (Spec, page 1, line 31 to page 2, line 2).
5. The term "non-sticky" means that the polymer particles do not stick to each other and remain free with respect to each other (Spec, page 2, lines 4-5).

6. PPTA aramid crumbs derived only from PPD and TDC monomers (herein referred to as "non-DAPBI PPTA aramid crumbs") are known (Spec, page 2, lines 10-11).
7. Prior to the present invention, a DAPBI monomer was added to PPD and TDC monomers to keep the DAPBI PPTA aramid polymer in a liquid solution, whereby the DAPBI PPTA aramid solution could then be further processed; DAPBI PPTA crumbs were heretofore previously unknown (Spec, page 2, lines 15-17).
8. The present inventors determined how to prepare a PPTA aramid crumb containing DAPBI ("DAPBI PPTA aramid crumb") when using PPD, DAPBI and TDC monomers, based upon specific materials and amounts, i.e., based upon the proper ratio of PPD, DAPBI, TDC, solvent and calcium chloride (Spec, page 2, lines 17-19).
9. Failing to select the proper materials and proper ratio of PPD, TDC, DAPBI, solvent and calcium chloride does not result in a DAPBI PPTA crumb, but a solution (as in Fact 7), powder, paste or dough, all of which are ill-suited for being dissolved in sulfuric acid to form a spin dope, which can be further processed into fibers and films (Spec, page 3, lines 17-19).
10. The present inventors thus developed a method for forming a DAPBI PPTA crumb by copolymerizing: i) "a" mole % of PPD; ii) "b" mole % of DAPBI; and iii) 90-110 mole % of TDC in a mixture of N-methyl pyrrolidone

("NMP") and containing "c" wt.% of CaCl_2 , wherein (1) "c" is within the range from 1 to 20, (2) the ratio "a" : "b" ranges from 1 : 20 to 20 : 1, "a" + "b" is 100 mole%, (3) i), ii), and iii) together comprise 1-20 wt.% of the mixture, (4) the product "b"·"c" is at least 50 and less than 215, (5) the aromatic polyamide in the crumb has a relative viscosity η_{rel} of at least 4, (6) wherein the crumb is defined as non-sticky particles at least 95% of which have an average diameter of 0.7-15 mm and (7) wherein the crumb is formed directly by the copolymerizing (Claims 1 and 10; Spec, page 2, lines 21-32).

11. The present inventors also developed a DAPBI PPTA aramid crumb, obtained directly by copolymerizing PPD, DAPBI and TDC in a mixture of NMP and CaCl_2 , wherein the PPTA aramid crumb has a relative viscosity η_{rel} of at least 4, and wherein the PPTA aramid crumb is defined as non-sticky particles at least 95% of which have an average diameter of 0.7-15 mm. (Claim 2; Spec, page 2, lines 21-32).

12. None of the references cited by the Examiner disclose the method described in Fact 10 or the product described in Fact 11.

13. The Examiner rejected claims 1-5 and 10 under 35 U.S.C. §103(a) relying upon Vollbracht in view of Chernykh and in further view of Jung (June 15, 2009 Final Rejection ("FR"), page 2, paragraph 2).

14. The Examiner found that one having ordinary skill in the art would have found it obvious to have used the DAPBI monomer described in Chernykh and

Jung in Vollbracht's polyamide and achieved a crumb (FR, page 5, paragraph 1).

15. Appellant disagrees with the Examiner's findings in Fact 14.

16. The Examiner acknowledges that Vollbracht does not describe (1) the use of DAPBI monomer or (2) the ratios of DAPBI to PPD and TDC, which are suitable for crumb formation (FR, page 3, paragraph 1).

17. Vollbracht describes a method of producing a non-DAPBI PPTA crumb using only PPD monomers and TDC monomers in a NMP/CaCl₂ solvent/salt combination (Vollbracht, col. 1, lines 53-60 and the Abstract).

18. The method described in Vollbracht is an example of known prior art (Fact 7).

19. Vollbracht does not describe replacing a portion of the PPD with DAPBI, or describe how one might be able to form a crumb when using DAPBI as a monomer.

20. Vollbracht does not describe the following limitations recited in claims 1 and 10 (1) "b" mole percent of DAPBI, (2) the ratio of "a" mole percent of PPD to "b" mole percent of DAPBI is from 1 : 20 to 20 : 1, (3) "a" + "b" is 100 mole%, (4) the product of "b" and "c" weight percent of calcium chloride is at least 50 and less than 215, (5) a DAPBI PPTA aramid crumb having a relative viscosity of at least 4 and (6) the crumb formed directly by the copolymerizing of monomer.

21. The conditions for obtaining a crumb directly during the copolymerization are not described in Chernykh or Jung.
22. The Examiner acknowledges that Chernykh and Jung do not describe a crumb obtained directly by copolymerization (FR, page 4, paragraph 3 and page 6, paragraph 6).
23. Chernykh and Jung do not describe (1) a mole percent of PPD to "b" mole percent of DAPBI is from 1:20 to 20:1 (2) "a" + "b" is 100 mole%, (3) the product of "b" and "c" weight percent of calcium chloride is at least 50 and less than 215, (4) a DAPBI PPTA aramid crumb having a relative viscosity of at least 4, and (5) the crumb formed directly by the copolymerizing of monomer.
24. Chernykh discloses a method for producing an aramid solution comprising PPTA and 2-(p-phenylene) benzimidazole terephthalamide units by copolymerizing: (1) 10-80 mole percent of PPD, (2) 10-80 mole percent of DAPBI, and (3) 100 mole percent of TDC in a mixture of NMP and calcium chloride or lithium chloride (FR, page 3, paragraph 2, citing Chernykh, Example 1).
25. The composition and method disclosed in Example 1 of Chernykh are different from the composition and method recited in the present claims.
26. Example 1 of Chernykh discloses (1) a different phenylenediamine (Cl-PPD vs. PPD), (2) a different salt (lithium chloride vs. calcium chloride) and (3) a different solvent (DMAC v. NMP) than required in the independent

claims in order to obtain a solution from the composition. As such, the mole percent sum (a+b) using the data from Example 1 of Cherykh is on 60 mole percent. (Chernykh, Example 1).

27. Jung describes a method for producing a polyamide solution by the polycondensation of 100 mole percent TDC, 60 mole percent DAPBI ("b") and 40 mole percent PPD ("a") in NMP where the sum of "a" and "b" is 100 mole percent and the relative viscosity is 4.3 (FR, page 4, paragraph 1 and page 6, paragraph 4 (citing Jung, Example 8)).

28. Jung describes that metal halide solubility promoting additives (such as calcium chloride and lithium chloride) may be included in the polycondensation mixture in an amount between 0.2 and 10 weight percent of the total weight of the polycondensation solution (FR, page 4, paragraph 2 and page 6, paragraph 5 (citing Jung, col. 3, lines 19-35)).

29. Appellant argued and submitted evidence in the form of a Declaration by Mr. Johannes Bos, during prosecution to address why Jung cannot form a crumb (April 15, 2009 Rule 132 Declaration ("DEC")).

30. Mr. Bos has (1) a degree as an engineer from Hogere Technische School ("HTS") Chemistry and (2) a total of 19 years of work and research experience in the field of, among others, aramid polymers for use in fiber spinning processes (DEC, paragraphs 1 and 2).

31. Mr. Bos is a named inventor in multiple U.S. Patents and WIPO Patents, and is the named inventor in the application at issue in this appeal (DEC, paragraph 3).

32. In view of Facts 30 and Fact 31, Mr. Bos qualifies as one of ordinary skill in the art.

33. Mr. Bos concluded that Jung adds salts such as lithium chloride and calcium chloride to keep the polymer in solution if the solubility of the polymer is too low without the addition of such a salt (DEC, page 15, paragraph 1).

34. Mr. Bos concluded that if "a solubility promoting additive is added, it would have been expected, in view of Jung's statements, that the solubility of the polymer of [Jung's] Example 8 would have been enhanced further" (DEC, page 15, paragraphs 1 and 2).

35. One having ordinary skill in the art would not have added a solubility-promoting additive described in Jung to form a DAPBI PPTA aramid crumb because Jung merely describes the formation of PPTA polymer solution.

36. Vollbracht, Chernykh and Jung do not describe a DAPBI PPTA aramid crumb or the conditions necessary to form a DAPBI PPTA aramid crumb.

37. The Examiner found that "in all cases (with or without PPD or DARBI [i.e., DAPBI]) crumb formation strictly dictates by solubility of the polymer in a system." The Examiner further contends that "the crumb can be formed in both cases varying the solvent/non-solvent concentration depending on the

polymer structure" and a "polyamide, based on PPD can remain in the solution if the concentration of the precipitant is not sufficient" (FR, page 11, paragraph 2).

38. The Examiner found that DAPBI is a common monomer for a fiber-forming polymer and known for its "increased temperature application range" fire resistance and good mechanical properties (FR, page 4, paragraph 4).

39. The Examiner found that solubility of the DAPBI PPTA aramid depends on its structure and that relations between monomer content, calcium chloride and other technological parameters to obtain a crumb "can be adjusted by an artisan with [the] routine experimentation approach" (FR, page 4, paragraph 5).

40. Appellant disagrees with the Examiner's findings in Facts 37, 38 and 39.

41. The composition recited in claim 2 and the methods for forming the composition in claims 1 and 10 do not contain any precipitant (Fact 37) present when forming the DAPBI PPTA aramid.

42. No precipitation occurs in the forming the DAPBI PPTA aramid because the polymer is formed in the NMP/CaCl₂ solution after having obtained a certain chain length (i.e., molecular weight) and directly becomes a crumb.

43. This is confirmed by the fact that no additional non-solvent compound or precipitant is added to the reaction mixture in the present independent claims.

44. Mr. Bos' Declaration also rebutted the Examiner's findings in Facts 37, 38 and 39 by demonstrating the unpredictability in forming crumbs when using DAPBI, PPD and TDC.
45. Mr. Bos' Declaration contains seven sets of experiments (DEC, page 2, paragraph 2, page 10, paragraph 3 and page 14, paragraph 2 to page 16, paragraph 2).
46. The Fifth Set of Experiments demonstrated a comparison between Experiments 1 and 2 performed by the Research and Development Department of Teijin Aramid and reference data from various prior art (DEC, page 10, paragraph 1).
47. Experiments 1 and 2 and of the Fifth Set of Experiments demonstrated the conditions associated with forming a DAPBI PPTA aramid solutions. (DEC, page 10, Table 6).
48. Mr. Bos concluded that "[i]n view of the fact that those of ordinary skill in the art have not succeeded in forming PPTA crumbs using NMP/LiCl instead of NMP/CaCl₂, NMP/LiCl certainly will not give crumbs in DAPBI-containing aramid polymers. This has been confirmed by Example 4 of U.S. Patent No. 4,018,735 (Nakagawa), detailing the formation of a semi-transparent dope from the reaction of PPD, DAPBI, TDC and a NMP/LiCl solvent and by Example 34 of U.S. Patent No. 4,172,738 (Mera) disclosing the formation of a powder (see Table 8)" (DEC, page 11, paragraph 1).

49. The experiments (Experiments 1-7) of the Sixth Set of Experiments contain (1) amounts of PPD ranging from 30 to 90 mole%, (2) amounts of DAPBI ranging from 10 to 70 mole%, (3) amounts of calcium chloride ranging from 2.82 to 11.55 wt% and (4) a "b.c" value of at least 50 and less than 215 (DEC, page 12, paragraph 1 to page 13, paragraph 2).
50. The process conditions for the experiments described in the Sixth Set of Experiments are within the limits recited in the independent claims (DEC, page 13, paragraph 2).
51. Each of the experiments of the Sixth Set of Experiments formed a crumb (DEC, page 13, paragraph 3).
52. The Seventh Set of Experiments consisted of five comparative experiments (DEC, page 14, paragraph 1).
53. Comparative Experiments I-IV of the Seventh Set of Experiments were performed under the exact same conditions as Experiments 1-7 in the Sixth Set of Experiments of the Declaration, except that the amounts of DAPBI and of calcium chloride were changed (DEC, page 14, paragraph 1).
54. Comparative Experiment V of the Seventh Set of Experiments was performed under the exact conditions described in U.S. Patent No. 4,172,938, which is not cited by the Examiner (DEC, page 14, paragraph 1).

55. Comparative Experiments I-III and V possess a "b·c" value of 231.0, 219.6, 306.9 and 39.2, respectively, each of which is less than the "b·c" range recited in the independent claims.

56. Although Comparative Experiment IV possesses a "b·c" value with the range recited in the independent claims, Mr. Bos further explained in his Declaration that the difference the intrinsic viscosity between Experiment 6 in the Sixth Set of Experiments and Comparative Experiment IV is that Experiment 6 forms a crumb and Comparative Experiment IV forms a gel (DEC, page 14, paragraph 2).

57. Mr. Bos stated that "[i]f a gel is formed, then there is no further mixing and as a result, movement of the reactive ends of the polymer to each other is inhibited whereby the reaction rate considerably drops thereby resulting in low molecular weight material" (DEC, page 14, paragraph 2).

58. Mr. Bos concluded that "when the mole percent of DAPBI (b) changes within a range from 20 to 67 mole% and the weight percent of calcium chloride (c) in a range from 1.96 to 11.55 wt%, no crumb is formed, but a dough/paste, which are unsuitable for forming aramid fibers and films on a commercial scale" (DEC, page 14, paragraph 3).

59. None of the experiments in the Seventh Set of Experiments formed a crumb (DEC, page 14, paragraph 3 and page 14, Table 8).

60. Mr. Bos concluded that "[i]t can therefore be concluded that replacing a portion of the PPD monomer with DAPBI in NMP/CaCl₂ will not form a crumb under reaction conditions that are different from the conditions recited in claims 1 and 10 of the above-identified application. In other words, crumbs can be obtained using this system, but a very specific relationship between the amounts of DAPBI and of CaCl₂ is required, which conditions are recited in the claimed subject matter" (DEC, page 14, paragraph 1).

61. The evidence in Fifth, Sixth and Seventh Set of Experiments in the Declaration demonstrates that although the ranges of DAPBI and calcium chloride shown in Table 2 are similar to those shown in Table 1, the value of the mole percent of DAPBI "b" multiplied by the weight percent of calcium chloride "c" is essential in determining whether a crumb will be formed.

62. The Examiner's allegation of routine experimentation with respect to Fact 39 is incorrect because the evidence in the Fifth, Sixth and Seventh Set of Experiments demonstrates that the formation of a DAPBI PPTA aramid crumb is unpredictable and requires "a highly significant combination of properties" (i.e., amounts and conditions), none of which are described or suggested by Vollbracht, Chernykh or Jung.

63. The Examiner included various other random arguments in the Final Rejection (FR, page 3 and page 11).

64. The Examiner found that Chernykh discloses that lithium chloride and calcium chloride can be equally used to form an aromatic polyamide solution (FR, page 3, paragraph 4 and page 6, paragraph 1).
65. Appellant disagrees with the Examiner's findings in Fact 62.
66. Chernykh only relates to polymer solutions (Fact 24), where the addition of CaCl_2 or LiCl enhances the ability of the dissolving power of polymer.
67. Interchangeability between CaCl_2 or LiCl indicated in Chernykh only relates to conditions to obtain polycondensate solutions, not crumbs.
68. Mr. Bos confirmed in Experiments 1 and 2 of the Fifth Set of Experiments (Fact 46-48) that lithium chloride and calcium chloride behave differently when used to form an aramid crumb, and thus cannot be interchanged successfully as alleged by the Examiner.
69. The Examiner found that Table 2 of Chernykh describes the addition of 2.8-4.8 weight percent of calcium chloride or lithium chloride.
70. Table 2 of Chernykh – referring to Example 5 of Chernykh – describes a benzothiazole derivative (5-amino-2-p-aminophenylbenzothiazole (DAPBT)) with these materials instead of the benzimidazole derivative (i.e., DAPBI) recited in the independent claims.
71. Mr. Bos concluded that "making a prediction that the solvent/salt combination (NMP/CaCl_2) for the [DAPBT]-containing aramids of Example 5

would be applicable to [DAPBI]-containing aramids is clearly unsupported by the disclosure of Chernykh" (DEC, page 15, paragraph 3).

72. The Examiner found that that the evidence presented in Mr. Bos' Declaration was not commensurate with the scope of claims 1 and 2 because a "specific salt concentration" was allegedly not recited in these claims (FR, page 11, paragraph 3).

73. Appellant disagrees with the Examiner's findings in Fact 72.

74. The independent claims each recite that the amount of calcium chloride (designated in the independent claims as "c") is within a range of 1 to 20 weight percent (Claims 1, 2 and 10; Spec, lines 1-6).

75. The Sixth Set of Experiment in the Declaration describes calcium chloride salt in concentrations of 2.82 and 11.55 weight percent, which is within the range recited in claims 1, 2 and 10 (DEC, page 12, Table 7).

76. The Examiner rejected claims 2-5 under 35 U.S.C. §103(a) relying upon Chernykh or Jung in combination with Vollbracht or Encyclopedia (FR, page 5, paragraph 2).

77. Encyclopedia, similar to Vollbracht, merely discloses that the polymerization of PPD and TDC leads to a PPTA polyamide in the form of a crumb (Encyclopedia, page 565, lines 14-20).

78. Encyclopedia does not describe replacing a portion of the PPD with DAPBI.

IX. ARGUMENT

A. Introduction

1. Overview Of Rejections

The Examiner rejected (1) claims 1-5 and 10 under 35 U.S.C. §103(a) as allegedly having been obvious over Vollbracht in view of Chernykh and in further view of Jung (Facts 13-16); and (2) claims 2-5 under 35 U.S.C. §103(a) as allegedly having been obvious over Chernykh or Jung in view of Vollbracht or Encyclopedia (Facts 76-78).

2. Overview Of The Claimed Subject Matter

Appellant has determined the conditions (recited in independent claims 1, 2 and 10) and the materials necessary to form an aramid crumb directly during copolymerization when using DAPBI as a co-monomer with PPD and TDC. Facts 1-2, 4-5 and 10-11. DAPBI PPTA aramid crumbs are desirable because crumbs can be easily formed into a spin dope, which can be further processed into fibers and films. Fact 3. If a DAPBI PPTA aramid is formed by any method that does not fall within all of the conditions recited in the independent claims, the PPTA aramid is not derived in the form of a crumb, but is instead not derived at all, or is a liquid solution, paste, powder or the like. Fact 9.

B. Claims 1-5 And 10 Would Not Have Been Obvious Over Vollbracht in View of Chernykh And In Further View Of Jung

1. Vollbracht, Chernykh And Jung Do Not Describe The Limitations Recited In The Independent Claims

The cited references (Vollbracht, Chernykh and Jung), alone or in combination, fail to describe all of the limitations recited in the claims 1, 2 and 10. Fact 12. In view of (1) MPEP §2142.02 requiring “all words of a claim to be considered”, (2) MPEP § 2141.02 requiring consideration of the “[claimed] invention and prior art as a whole”, and (3) the Board of Patent Appeal and Interferences' recent confirmation that a proper, post-KSR obviousness determination still requires the Office make “a searching comparison of the claimed invention – including all its limitations – with the teaching of the prior art”, an obviousness rejection requires at least a suggestion of all of the claim elements. See also *In re Wada and Murphy*, Appeal 2007-3733, citing *In re Ochiai*, 71 F.3d 1565, 1572 (Fed. Cir. 1995) and *CFMT v. Yieldup Intern. Corp.*, 349 F.3d 1333, 1342 (Fed. Cir. 2003).

Because the cited references (Vollbracht, Chernykh and Jung) (Facts 12 and 17-28) fail to describe or suggest forming a DAPBI PPTA aramid crumb, the cited references cannot be found to have rendered obvious a DAPBI PPTA aramid crumb or method of making the same as recited in the independent claims.

2. Appellant Has Demonstrated That Obtaining A Crumb Directly During The Copolymerization of PPD, DAPBI And TDC Monomers Is Not A Matter Of Routine Experimentation

The Examiner's contention that the relationship between materials (i.e., monomers and salts) used to form a crumb can be adjusted by routine experimentation (Facts 37-39) is incorrect (Fact 40). The Examiner has incorrectly theorized how to form a DAPBI PPTA aramid crumb (Facts 41-43) and incorrectly analyzed the unexpected nature of forming a DAPBI PPTA aramid crumb (Facts 44-45). For at least these reasons, the Examiner's contention is factually incorrect and contrary to the evidence of record.

To support a conclusion that "routine experimentation" is within the teachings of the art, one must demonstrate that the Appellant's experimentation comes within or is described by the art. See *In re Fay*, 347, F.2d 597, 602 (C.C.P.A 1965).

In *Pfizer v. Apotex*, Pfizer argued that its production of amlodipine besylate was not a matter of "routine experimentation" or "routine testing." *Pfizer, Inc. v. Apotex, Inc.*, 82 USPQ 2d 1321, 1335 (Fed. Cir. 2007). The Federal Circuit disagreed and held via a two-prong analysis that because the prior art (1) provided a means (i.e., described the conditions) of creating acid addition salts and (2) provided a basis to conclude that the art was predictable and that there was nothing unexpected in the result, Pfizer merely had to verify these results by routine testing. *Id.* at 1335. The Federal Circuit reasoned that

the prior art demonstrated the specific characteristics of each salt, including solubility, pH, stability, etc., and that these experiments are often used by scientists to verify the physiochemical characteristics of each salt. *Id.* at 1335. *Pfizer* thus merely found the best salt among known salts by undertaking known (routine) testing in the art.

In *Sanofi v. Apotex*, the Federal Circuit upheld the district court's ruling that the separation of enantiomers was not "a simple or routine procedure and that success in separation, as well as the allocation of properties, was unpredictable." See *Sanofi Synthelabo Inc., v. Apotex, Inc.*, 89 USPQ2d 1370 (Fed. Cir. 2008).

**a. The Cited References Do Not Describe
The Conditions Necessary To
Form A DAPBI PPTA Aramid Crumb**

The present facts can be distinguished from the first prong of *Pfizer* (requiring a showing that the art provides a means for making the claimed material) because none of the cited references (Vollbracht, Chernykh or Jung) describe the conditions necessary (i.e., "the means of creating") a DAPBI PPTA aramid crumb using the specific monomers and salt/solvent system recited in the independent claims. Fact 12.

(1) Deficiencies Of Vollbracht

Although Vollbracht describes forming a non-DAPBI PPTA aramid crumb during the copolymerization (Fact 17), Vollbracht does not describe (1)

replacing a portion of the PPD with DAPBI, (2) how one might be able to form a crumb when using DAPBI as a monomer and (3) each of the limitations recited claims 1 and 10. Facts 17-20.

Vollbracht merely embodies the concept that non-DAPBI PPTA aramid crumbs are well known. Facts 7 and 18. One having ordinary skill in the art would understand that the conditions described therein are solely applicable to crumb formation without a DAPBI monomer (i.e., a non-DAPBI PPTA aramid crumb).

**(2) The Conditions For Obtaining A Crumb
Directly During The Copolymerization
Are Not Described In Chernykh Or Jung**

Chernykh and Jung fail to describe the conditions for obtaining a crumb directly during the copolymerization and for this reason fail to describe each of the limitations recited in the independent claims. Fact 21.

One having ordinary skill in the art, reviewing the teachings of Vollbracht, Chernykh and Jung, would have understood and expected that DAPBI PPTA aramid crumbs are not formed when PPD is replaced in part with DAPBI. This is because Chernykh and Jung, which include the use of DAPBI, describe that replacing a portion of the PPD with DAPBI results in a DAPBI PPTA aramid solution, not a DAPBI PPTA aramid crumb. Facts 21-28.

Chernykh and Jung do not describe any way to form a DAPBI PPTA aramid crumb, or provide any suggestion as to what of a myriad of possible conditions might be modified to possibly form a crumb. Facts 21 and 22.

**(3) Chernykh And Jung
Differ In Materials Used**

The composition and method disclosed in Example 1 of Chernykh are different from the composition and method recited in the present claims. Fact 26. Thus, the mole percent sum of the independent claims (a + b) using the data of Example 1 is only 60 mole percent (a=0, b=60) and not 100 mole percent as recited in the independent claims. Fact 26.

One having ordinary skill in the art would have understood that the conditions and materials described in Chernykh are only applicable to the formation of DAPBI PPTA aramid solutions, not crumbs.

Jung requires the addition of a solubility promoting additive. Fact 28. Appellant further submits that if one of ordinary skill in the art based his or her efforts to obtain a crumb solely on solubility (as alleged by the Examiner (Fact 37) for the reasons discussed below), such an individual would not have added a solubility-promoting additive described in Jung because, as addressed by Mr. Bos in the Declaration (Fact 33 and 34) Jung merely describes the formation of PPTA polymer solution. As such, if one having ordinary skill in the art sought a crumb, such an individual would omit the solubility promoting additive (Fact 28) and thus would omit any metal halide.

Appellant submits that an individual confronted with the teachings of Vollbracht and desiring to form a crumb would not have been provided with any reason or rationale from Jung to have used calcium chloride or lithium chloride to form a crumb. Fact 35.

(4) Conclusion

For the above reasons, the present facts clearly do not meet the first prong of Pfizer because the cited references do not describe the suitable conditions to form a DAPBI PPTA aramid crumb.

b. Mr. Bos' Rule 132 Declaration Demonstrates The Unpredictability Associated With The Formation Of A DAPBI PPTA Aramid Crumb

The present facts can also be distinguished from the second prong of *Pfizer* (requiring a showing that the invention was not a matter of routine experimentation) because the Rule 132 Declaration prepared by Mr. Bos (Facts 30-32) demonstrates how the formation of a DAPBI PPTA aramid crumb is unpredictable.

As detailed below, Appellant's efforts were not simply a matter of "routine experimentation." For this reason, the Examiner's allegation of routine experimentation (Fact 39) is factually incorrect.

(1) The Examiner Misconstrues The Subject Matter Recited In The Independent Claims

The Examiner bases the rejection on the theory that DAPBI PPTA crumb formation is strictly dictated by the solubility of the DAPBI PPTA polymer in a

system. Fact 39. This is entirely incorrect and demonstrates a fundamental misunderstanding regarding the claimed subject matter. Fact 40.

DAPBI PPTA crumb formation from a PPD, DAPBI, TDC and CaCl_2/NMP system is not exclusively, or even predominately, the result of the solubility of the DAPBI PPTA polymer formed from that system. Fact 9 and 10. Rather, a crumb is formed as a result of the specific materials used and the amounts of these materials, regardless of solubility. Fact 9 and Fact 10. The Examiner assumes that if one adjusts the solvent/non-solvent ratios, a DAPBI PPTA aramid crumb will precipitate out of solution. This is incorrect.

In reality, decreasing the solubility of the polymer in a mixture may result in the formation of undesirable solution, pastes or powders, not a DAPBI PPTA aramid crumb. Fact 9 and Fact 47.

Furthermore, the Examiner's contentions that a DAPBI PPTA aramid crumb can be formed by varying the solvent/salt concentration, and that the PPTA polymer can remain in solution if there is insufficient precipitant is factually unsupported and technologically incorrect. Fact 37 and Fact 40.

Crumb formation is clearly not dependent on precipitant presence because the composition recited in claim 2 and the methods for forming the composition in claims 1 and 10 do not contain any precipitant present when forming the PPTA polymer. Fact 41.

Moreover, no precipitation occurs in the system at all because no additional non-solvent compound or precipitant is added to the reaction mixture in the present independent claims. Fact 42.

For the above reasons, Appellant respectfully submit the Examiner has misconstrued the subject matter of the present independent claims.

**(2) Unpredictability In Forming
DAPBI PPTA Aramid Crumbs**

The Court of Claims and Patent Appeals held in *In re May* that "unpredictability may be established vis-à-vis a highly significant combination of properties." See *In re May*, 574 F.2d 1082, 1095 (CCPA 1978).

The Rule 132 Declaration prepared by Mr. Bos (Facts 30-32) has effectively demonstrated that forming a DAPBI PPTA aramid crumb directly during the copolymerization of PPD, DAPBI and TDC monomers is not a matter of routine experimentation because the result of replacing a portion of DAPBI with PPD is entirely unpredictable and requires "a highly significant combination of properties." Fact 44 and Fact 62.

Rather than being related to some single phenomenon, DAPBI PPTA crumb formation was in fact unpredictable. To demonstrate the unpredictability in obtaining a DAPBI PPTA aramid crumb, Mr. Bos performed three sets of experiments (i.e., the Fifth, Sixth and Seventh Set of Experiments in the Rule 132 Declaration) to show that DAPBI PPTA aramid crumbs are formed if the

components and conditions recited in the independent claims are satisfied.

Fact 45.

The Fifth Set of Experiments confirms the unpredictability of the art because those having ordinary skill in the art would not have succeeded in forming DAPBI PPTA aramid crumbs by simply manipulating the type of salt.

Facts 46-48.

The Sixth Set of Experiments confirms that specific amounts and specific materials are within the limits recited in claims 1, 2 and 10 and are unpredictably necessary to form a crumb. Facts 49, 50 and 51.

The Seventh Set of Experiments confirms DAPBI PPTA aramids may have an inherent viscosity that is similar to the DAPBI PPTA aramids prepared from the Sixth Set of Experiments, and yet still unpredictably not form a crumb. Facts 52-60.

As such, the extensive evidence presented by Appellant demonstrates that the formation of a DAPBI PPTA aramid crumb is unpredictable and requires specific materials, amounts and conditions, none of which are described or suggested by Vollbracht, Chernykh or Jung. Fact 61. For this reason, the Examiner's allegation of routine experimentation is incorrect.

Fact 62.

3. The Cited References Provided One Of Ordinary Skill In The Art With No Reasonable Expectation Of Success In Forming A DAPBI PPTA Aramid Crumb

Vollbracht, Chernykh and Jung would not have provided one of ordinary skill in the art with any reasonable expectation of success in forming a DAPBI PPTA aramid crumb. In *Pfizer v. Apotex*, the Federal Circuit noted that "obviousness cannot be avoided simply by a showing of some degree of unpredictability in the art so long as there was a reasonable probability of success." See *Pfizer, Inc. v. Apotex, Inc.*, 82 USPQ2d at 1332 (citing *In re Corkill*, 771 F.2d 1496, 1500 (Fed. Cir. 1985).

However, in the present case, Vollbracht, Chernykh, and Jung cannot be found to have rendered the present claims obvious because, as discussed above, these references are silent, and thus would not have provided one with any reasonable expectation of success, regarding the conditions necessary to form a DAPBI PPTA aramid crumb. Fact 12.

Furthermore, the evidence submitted by Mr. Bos in the Rule 132 Declaration further demonstrates the lack of a reasonable expectation of success in forming a DAPBI PPTA aramid because this evidence shows how a DAPBI PPTA aramid crumb is not formed by simply (1) adding DAPBI to a PPD and TDC monomers (i.e., the Sixth and Seventh Set of Experiments) or

(2) manipulating the solubility of the crumb (i.e., the Fifth Set of Experiments).

Facts 44-60.

As such, the cited references would not have provided one of ordinary skill in the art with any reasonable expectation of success in deriving the DAPBI PPTA aramid crumb of claim 2 or the method of forming the DAPBI PPTA aramid crumb recited in claims 1 and 10. In view of the above arguments and evidence, the present claims are not rendered obvious by Vollbracht, alone or in combination with Chernykh and Jung.

**4. The Examiner's Allegation That Chernykh
Describes The Equivalence Of LiCl And CaCl₂
Would Be Applicable To Crumb Formation Is Incorrect**

For completeness, Appellant will now address the other points argued by the Examiner. Fact 63.

The Examiner's allegation that Chernykh describes that the use of LiCl and CaCl₂ is equivalent is incorrect because Chernykh only relates to polymer solutions, where the addition of CaCl₂ or LiCl enhances the ability of the dissolving power of polymer. Fact 64-65. Any interchangeability between CaCl₂ or LiCl only relates to conditions to obtain polycondensate solutions in Chernykh, not crumbs. Fact 67. Moreover, as confirmed by Mr. Bos in the Rule 132 Declaration, particularly the Fifth Set of Experiments, lithium chloride and calcium chloride behave differently when used to form an aramid

crumb, and thus cannot be interchanged successfully as alleged by the Examiner. Facts 68-69.

5. The CaCl₂/NMP Salt/Solvent Combination Described In Table 2 Of Chernykh Is Directed To A Different Chemical Compound Than Recited In Claims 1 And 10

The CaCl₂/NMP salt/solvent combination described in Table 2 of Chernykh is directed to a benzothiazole derivative (Facts 69-71) and thus would not have provided one with any reason or rationale to have used the same salt/solvent combination in the benzimidazole derivative (DAPBI) described in claims 1 and 10.

In other words, the Examiner's contention that the solvent/salt combination (NMP/CaCl₂) used for forming DAPBT-containing aramids would apply to the DAPBI-containing aramids recited in the independent claims is entirely unsupported by the disclosure of Chernykh.

6. The Examiner's Allegation That The Evidence Presented In The Rule 132 Declaration Regarding The Alleged Equivalence of Lithium Chloride And Calcium Chloride Is Not Commensurate In Scope With The Independent Claims Is Incorrect

The Examiner's allegation that the evidence presented in the Rule 132 Declaration regarding the alleged equivalence of lithium chloride and calcium chloride is not commensurate in scope with the independent claims is incorrect. Facts 72-73. The Examiner reasons that claims 1 and 2 do not recite a specific salt concentration. Fact 72.

The independent claims each explicitly recite that the amount of calcium chloride (designated in the independent claims as "c") is within a range of 1 to 20 weight percent. Fact 74. Furthermore, the Sixth Set of Experiments in the Declaration state that the amount of calcium chloride used therein was between 2.82 and 11.55 weight percent. Fact 75. As such, the Examiner's dismissal of the evidence in the Rule 132 Declaration as allegedly not being commensurate with the scope of the independent claims is incorrect.

7. Conclusion

As such, none of the references describe (1) the composition in crumb form as recited in claim 2 or (2) the method to make the composition in crumb form as recited in claims 1 and 10. In view of the above arguments, Appellant respectfully requests withdrawal of the 35 U.S.C. §103(a) rejection.

C. Claims 2-5 Would Not Have Been Obvious Over Chernykh Or Jung in View of Vollbracht Or Encyclopedia

For the reasons described above, Chernykh, Jung and Vollbracht do not describe the subject matter recited in claim 2. Fact 78. Encyclopedia does not remedy the deficiencies of these references. Fact 78. Encyclopedia, cited by the Examiner as allegedly suggesting the composition of claims 2-5, actually illustrates why the references would not have led one of ordinary skill in the art to the present claims.

Encyclopedia, similar to Vollbracht, merely discloses that the copolymerization of PPD and TDC leads to a PPTA polyamide in the form of a

crumb. Fact 77. However, Encyclopedia does not describe replacing a portion of the PPD with DAPBI. Fact 78. No specific requirements to make a crumb must be met when using PPD alone as in Encyclopedia as illustrated in Vollbracht. Facts 16-19.

Thus, it would not have been obvious to one of ordinary skill in the art to have combined the teachings of Chernykh with Jung to form the crumb obtained in Vollbracht and Encyclopedia using a different composition.

Furthermore, Chernykh or Jung do not disclose a crumb, as the processes disclosed in Chernykh or Jung directly process a polymer from a solution. Fact 22. Nothing in any of the references would have led one of ordinary skill in the art to have modified the compositions described in Vollbracht, Chernykh or Jung to the composition of claim 2 with any reasonable expectation of success in being able to obtain the composition in crumb form.

Withdrawal of the rejection is respectfully requested.

For all of the reasons discussed above, it is respectfully submitted that the rejections are in error and that claims 1-5 and 10 are in condition for allowance. For all of the above reasons, Appellant respectfully request this Honorable Board to reverse the rejections of claims 1-5 and 10.

Respectfully submitted,

A handwritten signature in black ink, appearing to read 'William P. Berridge', written over a horizontal line.

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X. APPENDIX A - CLAIMS SECTION

1. (Rejected) A method for obtaining an aromatic polyamide containing para-phenylene terephthalamide and 2-(p-phenylene)benzimidazole terephthalamide units, in the form of a crumb, by copolymerizing:

 i) a mole % of para-phenylenediamine;

 ii) b mole % of 5(6)-amino-2-(p-aminophenyl)benzimidazole; and

 iii) 90-110 mole% of terephthaloyl dichloride in a mixture of N-methyl pyrrolidone and containing c wt.% of calcium chloride,

 wherein c is within the range from 1 to 20, and wherein the ratio a : b ranges from 1 : 20 to 20 : 1, a + b is 100 mole%, and i), ii), and iii) together comprise 1-20 wt.% of the mixture,

 wherein the product b.c is at least 50 and less than 215 and the aromatic polyamide in the crumb has a relative viscosity η_{rel} of at least 4,

 wherein the crumb is defined as non-sticky particles at least 95% of which having an average diameter of 0.7-15 mm, and

 wherein the crumb is formed directly by the copolymerizing.

2. (Rejected) An aromatic polyamide containing para-phenylene terephthalamide and 2-(p-phenylene)benzimidazole terephthalamide units, in the form of a crumb, obtained directly by copolymerizing para-phenylenediamine; 5(6)-amino-2-(aminophenyl)benzimidazole; and

terephthaloyl dichloride in a mixture of N-methyl pyrrolidone and calcium chloride,

wherein the aromatic polyamide in the crumb has a relative viscosity η_{rel} of at least 4, and

wherein the crumb is defined as non-sticky particles at least 95% of which having an average diameter of 0.7-15 mm.

3. (Rejected) The aromatic polyamide of claim 2, wherein the aromatic polyamide in the crumb has a relative viscosity η_{rel} between 4 and 7.

4. (Rejected) A method for making purified aromatic polyamide by coagulating and washing the crumb of claim 2 in water, followed by a drying step.

5. (Rejected) A method for making purified aromatic polyamide by coagulating and washing the crumb of claim 3 in water, followed by a drying step.

6. (Withdrawn) A method for obtaining a composition comprising the purified aromatic polyamide obtained by the method of claim 4, the method comprising:

dissolving the purified aromatic polyamide in the form of a crumb in a solvent.

7. (Withdrawn) The method of claim 6, wherein the solvent is selected from the group consisting of sulfuric acid, N-methyl pyrrolidone, dimethylacetamide and combinations thereof.

8. (Withdrawn) A method for obtaining a composition comprising the purified aromatic polyamide obtained by the method of claim 5, the method comprising:

dissolving the purified aromatic polyamide in the form of a crumb in a solvent.

9. (Withdrawn) The method of claim 8, wherein the solvent is selected from the group consisting of sulfuric acid, N-methyl pyrrolidone, dimethylacetamide and combinations thereof.

10. (Rejected) A method for obtaining an aromatic polyamide containing para-phenylene terephthalamide and 2-(p-phenylene)benzimidazole terephthalamide units, in the form of a crumb, the method consisting of copolymerizing:

- i) a mole % of para-phenylenediamine;
- ii) b mole % of 5(6)-amino-2-(p-aminophenyl)benzimidazole; and
- iii) 90-110 mole% of terephthaloyl dichloride in a mixture of N-methyl pyrrolidone and containing c wt.% of calcium chloride,

wherein c is within the range from 1 to 20, and wherein the ratio $a : b$ ranges from 1 : 20 to 20 : 1, $a + b$ is 100 mole%, and i), ii), and iii) together comprise 1-20 wt.% of the mixture,

wherein the product $b.c$ is at least 50 and less than 215 and the aromatic polyamide in the crumb has a relative viscosity η_{rel} of at least 4, and

wherein the crumb is defined as non-sticky particles at least 95% of which having an average diameter of 0.7-15 mm.

**XI. APPENDIX B - CLAIM SUPPORT
AND DRAWING ANALYSIS SECTION**

1. A method for obtaining an aromatic polyamide {page 1, line 3} containing para-phenylene terephthalamide and 2-(p-phenylene)benzimidazole terephthalamide units {page 1, lines 3-5}, in the form of a crumb {page 2 lines 2-9}, by copolymerizing {page 1, line 4}:

- i) a mole % of para-phenylenediamine {page 2, line 26};
- ii) b mole % of 5(6)-amino-2-(p-aminophenyl)benzimidazole {page 2, line 27}; and
- iii) 90-110 mole% of terephthaloyl dichloride in a mixture of N-methyl pyrrolidone and containing c wt.% of calcium chloride {page 2, lines 28-29},

wherein c is within the range from 1 to 20 {page 2, lines 29-30}, and wherein the ratio a : b ranges from 1 : 20 to 20 : 1, a + b is 100 mole%, and i), ii), and iii) together comprise 1-20 wt.% of the mixture {page 2, lines 30-31},

wherein the product b.c is at least 50 and less than 215 {page 2, line 32} and the aromatic polyamide in the crumb has a relative viscosity η_{rel} of at least 4 {page 32, line 32 to page 3, line 1},

wherein the crumb is defined as non-sticky particles at least 95% of which having an average diameter of 0.7-15 mm {page 2, lines 7-9; page 3, lines 1-2}, and

wherein the crumb is formed directly by the copolymerizing {**page 1, lines 30-31; page 2, lines 13-17; and page 3, lines 20-37**}.

2. An aromatic polyamide {**page 1, line 3**} containing para-phenylene terephthalamide and 2-(p-phenylene)benzimidazole terephthalamide units {**page 1, lines 3-5**}, in the form of a crumb {**page 2 lines 2-9**}, obtained directly {**page 1, lines 30-31; page 2, lines 13-17; and page 3, lines 20-37**} by copolymerizing para-phenylenediamine {**page 2, line 26**}; 5(6)-amino-2-(aminophenyl)benzimidazole {**page 2, line 27**}; and terephthaloyl dichloride in a mixture of N-methyl pyrrolidone and calcium chloride {**page 2, lines 28-29**},

wherein the aromatic polyamide in the crumb has a relative viscosity η_{rel} of at least 4 {**page 32, line 32 to page 3, line 1**}, and

wherein the crumb is defined as non-sticky particles at least 95% of which having an average diameter of 0.7-15 mm {**page 2, lines 7-9; page 3, lines 1-2**}.

10. A method for obtaining an aromatic polyamide {**page 1, line 3**} containing para-phenylene terephthalamide and 2-(p-phenylene)benzimidazole terephthalamide units {**page 1, lines 3-5**}, in the form of a crumb {**page 2 lines 2-9**}, the method consisting of copolymerizing {**page 1, line 4**}:

i) a mole % of para-phenylenediamine {**page 2, line 26**};

ii) b mole % of 5(6)-amino-2-(p-aminophenyl)benzimidazole
{**page 2, line 27**}; and

iii) 90-110 mole% of terephthaloyl dichloride in a mixture of N-methyl pyrrolidone and containing c wt.% of calcium chloride {**page 2, lines 28-29**},

wherein c is within the range from 1 to 20 {**page 2, lines 29-30**},
and wherein the ratio a : b ranges from 1 : 20 to 20 : 1, a + b is 100 mole%, and
i), ii), and iii) together comprise 1-20 wt.% of the mixture {**page 2, lines 30-31**},

wherein the product b.c is at least 50 and less than 215 {**page 2, line 32**} and the aromatic polyamide in the crumb has a relative viscosity η_{rel} of at least 4 {**page 32, line 32 to page 3, line 1**}, and

wherein the crumb is defined as non-sticky particles at least 95% of which having an average diameter of 0.7-15 mm {**page 2, lines 7-9; page 3, lines 1-2**}.

**XII. APPENDIX C - MEANS OR STEP PLUS
FUNCTION ANALYSIS SECTION**

NONE

XIII. APPENDIX D - EVIDENCE SECTION

A copy of each of the following items of evidence relied on by the Appellant and/or the Examiner in this appeal is attached:

Table of Contents

	<u>Page</u>
Request for Reconsideration filed April 15, 2009	49
Rule 132 Declaration filed April 15, 2009	62
Final Rejection mailed June 15, 2009	86
Specification of Application No. 10/580,018, filed May 19, 2006	101

PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of

Johannes BOS

Group Art Unit: 1796

Application No.: 10/580,018

Examiner: G. LISTVOYB

Filed: May 19, 2006

Docket No.: 127898

For: PROCESS FOR MAKING DAPBI-CONTAINING ARAMID CRUMBS

REQUEST FOR RECONSIDERATION

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

In reply to the November 17, 2008, Office Action, reconsideration of the rejection is respectfully requested in light of the following remarks.

REMARKS

Claims 1-10 are pending in this application. Claims 6-9 are currently withdrawn.

Applicant respectfully requests reconsideration of the pending claims.

I. Interview

The courtesies extended to Applicant's representative by Examiner Listvoyb at the interview held February 11, 2009, are appreciated. The reasons presented at the interview as warranting favorable action are incorporated into the remarks below, which constitute Applicant's record of the interview.

II. Rejections Under 35 U.S.C. §103(a)

A. Claims 1-5 and 10

Claims 1-5 and 10 are rejected under 35 U.S.C. §103(a) as allegedly being unpatentable over U.S. Patent No. 4,308,374 ("Vollbracht") in view of RU 2017866 ("Chernykh") and in further view of U.S. Patent No. 5,646,234 ("Jung"). Applicant respectfully traverses this rejection.

The Patent Office alleges that it would have been obvious to one of ordinary skill in the art to have taken the polymers disclosed in Chernykh and Jung and expanded the applicability of these polymers using the technology disclosed in Vollbracht. See Office Action, page 5. Applicant respectfully disagrees.

Specifically, although Vollbracht describes forming a crumb during the copolymerization, Vollbracht describes forming a crumb using entirely different reaction materials from the materials recited in the present claims. Furthermore, Chernykh or Jung do not disclose a crumb, as the processes disclosed in Chernykh or Jung directly process a polymer from a solution. Nothing in any of the references would have led one of ordinary skill in the art to have modified the compositions described in Vollbracht, Chernykh or Jung to the composition of claim 2 with any reasonable expectation of success in being able to

obtain the composition in crumb form. Nor would the references have led one of ordinary skill in the art to the process of claims 1 and 10, as the references fail to describe how to obtain the composition in crumb form as recited in claim 2.

1. Vollbracht

Vollbracht describes a method of producing an aromatic polyamide ("PPTA") crumb using para-phenylenediamine ("PPD") monomers and terephthaloyl dichloride ("TDC") monomers in a N-methyl pyrrolidone/calcium chloride ("NMP/CaCl₂") solvent/salt combination. See Vollbracht, col. 1, lines 53-60 and the Abstract. PPTA crumbs are known and highly desired as the purification of crumbs is relatively easy and the processability is high. See page 2, lines 10-11 of the specification. As described in the First Set of Experiments in the attached Rule 132 Declaration – which represent the embodiments described in Vollbracht – varying the concentration of PPD and TDC monomers and the weight percentage of CaCl₂ produces a PPTA crumb with an inherent viscosity (η_{inh}) that was always greater than 3.

Vollbracht does not describe replacing a portion of the PPD with 5(6)-amino-2-(p-aminophenyl)-benzimidazole ("DAPBI"). Specifically, Applicant alone has determined that if a portion of the PPD is replaced by DAPBI, the proper ratio of PPD, DAPBI and calcium chloride must be used or the aramid polymer will not form a crumb, but a sticky paste, dough or powder. See page 2, lines 21-22 of the specification. As such, none of the references describe (1) the modification of PPD with DAPBI to form a crumb and (2) the amounts of PPD, DAPBI and calcium chloride required to obtain a crumb. Thus, it would not have been obvious to one of ordinary skill in the art to have combined the teachings of Chernykh with Jung to form the crumb obtained in Vollbracht using a different composition.

2. Chernykh

The Patent Office alleges that Chernykh discloses a method for producing an aromatic polyamide (aramid) comprising para-phenylene terephthalamide and 2-(p-phenylene) benzimidazole terephthalamide units by copolymerizing: (1) 10-80 mole percent of PPD, (2) 10-80 mole percent of DAPBI, and (3) 100 mole percent of TDC in a mixture of NMP and calcium chloride or lithium chloride. See Office Action, page 4, citing Chernykh, Example 1. Applicant respectfully disagrees.

As admitted by the Patent Office, Chernykh does not describe a crumb obtained directly by copolymerization because the processes described in Chernykh maintain the reaction in solution after polymerization and directly use the solution to form a spin dope. See Office Action, page 5.

a) **Chernykh Would Not Have Provided One With Any Reason or Rationale To Have Replaced A Portion of PPD With DAPBI And Form A Crumb**

One having ordinary skill in the art, starting with the teachings of Vollbracht, would have understood that crumbs are not formed when PPD is replaced by a portion of DAPBI. Such a person, in looking for a solution to this problem, would not have been provided with any reason or rationale from Chernykh or Vollbracht to have combined these references. Chernykh's description of forming a solution by substituting a portion of PPD with DAPBI would have directed one to have believed that a crumb could not have been formed by this substitution.

Furthermore, Chernykh does not describe the conditions as presently recited in claims 1, 2 and 10. Example 1 of Chernykh discloses a method for producing an aromatic polyamide by copolymerizing: (1) 40 mole percent of 2-chloro-para-phenylenediamine (Cl-PPD); (2) 60 mole percent of DAPBI and (3) 100 mole percent of TDC in a mixture of anhydrous N,N-dimethylacetamide ("DMAc") and lithium chloride.

In contrast, claim 2 of the present application recites an aromatic polyamide containing para-phenylene terephthalamide and 2-(p-phenylene)benzimidazole terephthalamide units, in the form of a crumb, obtained directly by copolymerizing para-phenylenediamine; 5(6)-amino-2-(aminophenyl)benzimidazole; and terephthaloyl dichloride in a mixture of N-methyl pyrrolidone and calcium chloride, wherein the aromatic polyamide in the crumb has a relative viscosity η_{rel} of at least 4, and wherein the crumb is defined as non-sticky particles at least 95% of which having an average diameter of 0.7-15 mm.

Also, claim 1 of the present application recites a method for obtaining an aromatic polyamide containing para-phenylene terephthalamide and 2-(p-phenylene)benzimidazole terephthalamide units, in the form of a crumb, by copolymerizing: i) a mole % of para-phenylenediamine; ii) b mole % of 5(6)-amino-2-(p-aminophenyl)benzimidazole; and iii) 90-110 mole% of terephthaloyl dichloride in a mixture of N-methyl pyrrolidone and containing c wt.% of calcium chloride, wherein c is within the range from 1 to 20, and wherein the ratio a : b ranges from 1 : 20 to 20 : 1, a + b is 100 mole%, and i), ii), and iii) together comprise 1-20 wt.% of the mixture, wherein the product b.c is at least 50 and less than 215 and the aromatic polyamide in the crumb has a relative viscosity η_{rel} of at least 4, wherein the crumb is defined as non-sticky particles at least 95% of which having an average diameter of 0.7-15 mm, and wherein the crumb is formed directly by the copolymerizing.

Thus, the composition and method disclosed in Example 1 of Chernykh are different than the composition and method recited in the present claims. Example 1 of Chernykh discloses (1) a different phenylenediamine (Cl-PPD vs. PPD), (2) a different salt (lithium chloride v. calcium chloride) and (3) a different solvent (DMAC v. NMP) than required in claim 2 in order to obtain a crumb form of the composition. As such, one having ordinary skill in the art would have been required to have performed numerous substitutions to have arrived at the subject matter recited in claim 1.

**b) The Conditions For Obtaining A Crumb Directly During
The Copolymerization Are Not Described In Chernykh**

The copolymerization of PPD and TDC leads to a PPTA crumb. However, the present application embodies the scenario when a portion of PPD has been replaced with DAPBI. Prior to the present application, such a replacement formed a powder compound or a gel compound, which has been a serious problem for large-scale production of a DAPBI-containing aramid.

As described in the Fifth Set of Experiments in the Rule 132 Declaration, forming a PPTA aramid crumb using PPD, DAPBI and TDC monomers requires a specific set of conditions. The Fifth Set of Experiments includes Applicants' reproductions of Examples 1, 4 and 7 of Chernykh. As shown in Table 6 of the Rule 132 Declaration, Examples 1, 4 and 7 describe the formation of a PPTA polymer solution using (1) Cl-PPD or PPD, (2) DAPBI and (2) DMAc alone or in combination with LiCl. As such, this evidence confirms that the Examples of Chernykh do not form a crumb. Those having ordinary skill in the art would not have succeeded in forming PPTA crumbs using the NMP/LiCl described in Chernykh instead of the NMP/CaCl₂ described in the present claims.

Such a conclusion has been confirmed (1) by Example 4 of U.S. Patent No. 4,018,735 (Nakagawa) detailing the formation of a semi-transparent aramid dope (i.e., a solution) from the reaction of PPD, DAPBI, and TDC monomers in a LiCl/NMP salt/solvent combination and (2) by Example 34 of U.S. Patent No. 4,172,938 ("Mera") disclosing the formation of a powder because of the b.c value is not within the range recited in claims 1 and 10.

To demonstrate the difficulty in obtaining a crumb from a DAPBI polymer, Applicant performed two additional sets of experiments to show that crumbs are formed if the components and conditions recited in present claims 1 and 10 are satisfied. These two sets of experiments are characterized as the Sixth and Seventh Set of Experiments in the Rule 132

Declaration, wherein the results of these experiments are illustrated in Table 7 and Table 8 of the Rule 132 Declaration, respectively. The Sixth Set of Experiments is represented as Examples 1-7 as described on page 5 of the present specification and the Seventh Set of Experiments is represented as Comparative Examples 1-5 also described on page 5 of the present specification.

As shown in Table 7 of the Rule 132 Declaration, the experiments (Experiments 1-7) of the Sixth Set of Experiments each contain (1) amounts of PPD ranging from 30 to 90 mole%, (2) amounts of DAPBI ranging from 10 to 70 mole% and (3) amounts of calcium chloride ranging from 2.82 to 11.55 wt%. Furthermore, each of the compositions for these experiments formed a crumb directly during copolymerization due to each composition possessing an inherent viscosity of at least 4.0.

Furthermore, shown in Table 8 of the Rule 132 Declaration, if one alters (1) the mole% of DAPBI (b) in a range from 20 to 67 mole% and (2) the weight percent of calcium chloride (c) in a range from 1.96 to 11.55 wt%, then a crumb will not be formed, but a dough/paste, gel or powder will be formed. Such products are less suitable for forming aramid fibers and films in a commercial scale production. This evidence demonstrates that although the ranges of DAPBI and calcium chloride shown in Table 8 are similar to those shown in Table 7, the value of the mole percent of DAPBI (b) multiplied by the weight percent of calcium chloride (c) is essential in determining whether a crumb will be formed.

As such, one will conclude that replacing a portion of the PPD monomer with DAPBI monomer in NMP/CaCl₂ will not form a crumb under reaction conditions that are different from the conditions recited in claims 1 and 10 of the above-identified application. In other words, crumbs obtained using these materials require a very specific relationship between the amounts of DAPBI and of CaCl₂, none of which are described in Chernykh.

c) The Patent Office's Allegation That Chernykh Describes The Equivalence Of LiCl And CaCl₂ Would Be Applicable To Crumb Formation Is Incorrect

The Patent Office alleges that Chernykh describes that the use of LiCl and CaCl₂ is equivalent. See Office Action, page 4. Chernykh mentions the use of "CaCl₂ or LiCl", but does not reference what the term "or" actually means. Chernykh's use of the word "or" does not mean that if one replaced CaCl₂ with LiCl, all properties and effects of the polymer remain the same. For example, the viscosity of the polymer solution, the relative viscosity of the polymer, etc., can each be different. Thus, the term "or" in Chernykh cannot be interpreted as meaning that both salts can be used interchangeably without altering the properties of the polymer.

As described above, Chernykh only relates to polymer solutions, where the addition of CaCl₂ or LiCl enhances the ability of the dissolving power of polymer. As such, any interchangeability between CaCl₂ or LiCl only relates to conditions to obtain polycondensate solutions. But lithium chloride and calcium chloride behave differently when used to form an aramid crumb, and thus cannot be interchanged successfully as allegedly by the Patent Office.

Attached herewith is a Rule 132 Declaration that contains four sets of polymerization experiments to show calcium chloride and lithium chloride are not equivalent as compounds for forming an aramid crumb.

The First Set of Experiments in the attached Rule 132 Declaration describe varying the concentration of PPD and TDC monomers and the weight percentage of CaCl₂ in the NMP/CaCl₂ used to form the PPTA polymer. As shown in Table 1 of the Rule 132 Declaration, this variance always resulted in a PPTA polymer with an inherent viscosity (η_{inh}) greater than 3 and thus the variance did not affect the formation of a PPTA crumb.

The Second Set of Experiments in the Rule 132 Declaration was performed in the exact same manner as the First Set of Experiments, except that CaCl₂ was replaced with other

salts, including lithium chloride. As shown in Table 2, the addition of LiCl only resulted in an inherent viscosity of 0.30, whereas the addition of calcium chloride (as shown in Table 1) unexpectedly achieved a substantially greater inherent viscosity to enable crumb formation.

The Third Set of Experiments in the Rule 132 Declaration describes that crumb formation also depends upon (1) the monomer concentration and (2) salt concentration. As shown in Table 3 of the Declaration, the final products of the Third Set of Experiments had an η_{inh} between 0.38 and 0.63 and thus formed a turbid yellow to a brown syrupy mass instead of a crumb.

The Fourth Set of Experiments in the Rule 132 Declaration was performed in the exact same manner as the First and Second Set of Experiments, except that the LiCl and CaCl₂ salts were combined with a dimethyl acetamide (DMAc) or NMP solvent. As shown in Table 4 of the Rule 132 Declaration, the NMP/CaCl₂ solvent/salt combination had an η_{inh} value of 4.31 and thus formed a crumb. But the remaining solvent/salt combinations, including (1) LiCl/DMAc (described in Example 1 of Chernykh), (2) CaCl₂/DMAc and (3) LiCl/NMP, each formed a solution comprised of low molecular weight materials and possessed too low of an inherent viscosity to form a crumb directly during the copolymerization.

The four sets of Experiments in the Rule 132 Declaration demonstrate that the formation of PPTA from TDC and PPD monomers in a CaCl₂/NMP solvent/salt combination renders a sufficient inherent viscosity to form a PPTA crumb. Yet the replacement of the CaCl₂ solvent with a LiCl solvent does not render a sufficient inherent viscosity to form a PPTA crumb. As such, the four sets of experiments in the Rule 132 Declaration confirm that LiCl and CaCl₂ are not equivalent and cannot be used interchangeably when the desired final product is a PPTA crumb.

d) The CaCl₂/NMP Salt/Solvent Combination Described In Table 2 Of Chernykh Is Directed To A Different Chemical Compound Than Recited In Claims 1 And 10

The CaCl₂/NMP salt/solvent combination described in Table 2 of Chernykh is directed to a benzothiazole derivative and thus would not have provided one with any reason or rationale to have used the same salt/solvent combination in the benzimidazole derivative described in claims 1 and 10.

The Patent Office alleges that Table 2 of Chernykh describes the combination of NMP and CaCl₂. See Office Action, page 4. Applicants respectfully disagree.

Table 2 of Chernykh – referring to Example 5 of Chernykh – describes a benzothiazole derivative (5-amino-2-p-aminophenylbenzothiazole (DAPBT)) instead of the benzimidazole derivative (i.e., DAPBI) recited in claims 1, 2 and 10. However, the present claims are limited to DAPBI-containing aramids (not DAPBT aramids). Furthermore, the thiazole compound described in Chernykh possesses a much different functionality and/or characteristics than the imidazole compounds recited in the present claims.

As such, the Patent Office's contention that the solvent/salt combination (NMP/CaCl₂) used for forming of DABPT-containing aramids would not apply to the DAPBI-containing aramids recited in claim 1-10. Such a contention is entirely unsupported by the disclosure of Chernykh.

3. Jung

The Patent Office alleges that Jung discloses a method for producing a polyamide by the polycondensation of 100 mole percent TDC, 60 mole percent DAPBI (a) and 40 mole percent PPD (b) in NMP where the sum of a and b is 100 mole percent and relative viscosity is 4.3. See Office Action, page 4, citing Jung, Example 8. Applicant respectfully disagrees.

Jung also fails to describe the composition of claim 2, a composition in crumb form, or how to obtain the composition of claim 2 in crumb form by the use of the process (solvents and amounts of materials) of claim 1 and claim 10. According to processes described in claims 1 and 10, the polymer should not remain dissolved in the solvent (i.e., a solution) nor precipitate from the solvent. In contrast, the processes described in claims 1 and 10 should form a dough or "liquid cheese-like" material, which becomes a crumb upon further stirring.

The Patent Office alleges that Jung describes that use of a solubility promoting additive such as CaCl_2 can be added to the polycondensation mixture. See Office Action, page 5. Further, as admitted by the Patent Office, Jung does not describe the formation of a crumb directly by copolymerization, but the formation of PPTA polymer solutions. See Office Action, page 5.

Because Jung merely describes the formation of PPTA polymer solutions, one having ordinary skill in the art, wishing to obtain a crumb during copolymerization, would not have added a solubility-promoting additive. Such a person would expect that the inclusion of solubility promoting additive would form a soluble polymer (i.e., a solution) rather than a sticky paste that converts to an insoluble crumb upon additional stirring. As such, an individual confronted with the teachings of Vollbracht and desiring to form a crumb would not have been provided with any reason or rationale from Jung, but instead would have searched for another solvent that does not dissolve the final PPTA polymer.

As such, one having ordinary skill in the art having the desire to form a crumb directly during copolymerization, would not have added an alkali metal salt or an alkaline earth metal salt because Jung (and Chernykh) describe that such addition increases the solubility of the polymer to form a solution, not a crumb.

4. Conclusion

As such, none of the references describe (1) the composition in crumb form as recited in claim 2 or (2) the method to make the composition in crumb form as recited in claims 1 and 10. In view of the above arguments, Applicant respectfully requests withdrawal of the above 35 U.S.C. §103(a) rejection.

B. Claims 2-5

Claims 2-5 are rejected under 35 U.S.C. §103(a) as allegedly being unpatentable over Chernykh in further view of Jung and in still further view of Vollbracht in combination with Encyclopedia of Pol. Sci. and Tech, Polyamides, vol. 3, p. 565-567 ("Encyclopedia").

Applicant respectfully traverses this rejection.

For the reasons described above, Chernykh, Jung and Vollbracht do not describe the subject matter recited in claim 2. Encyclopedia does not remedy the deficiencies of these references. Encyclopedia, cited by the Patent Office as allegedly suggesting the composition of claims 2-5, actually illustrates why the references would not have led one of ordinary skill in the art to the present claims.

Encyclopedia, similar to Vollbracht, merely discloses that the copolymerization of PPD and TDC leads to a PPTA polyamide in the form of a crumb. See Encyclopedia, page 565, lines 14-20. However, Encyclopedia does not describe replacing a portion of the PPD with DAPBI. No specific requirements to make a crumb must be met when using PPD alone as in Encyclopedia as illustrated above in the First Set of Experiments in the Rule 132 Declaration.

Thus, it would not have been obvious to one of ordinary skill in the art to have combined the teachings of Chernykh with Jung to form the crumb obtained in Vollbracht and Encyclopedia using a different composition.

Withdrawal of the rejection is respectfully requested.

III. Rejoinder

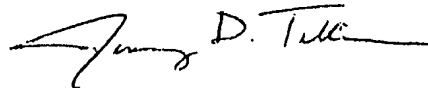
In view of the foregoing amendments and arguments, Applicant respectfully requests that upon allowance of claims 1-5 and 10, claims 6-9 be rejoined with the present application and similarly allowed.

IV. Conclusion

In view of the foregoing, it is respectfully submitted that this application is in condition for allowance. Favorable reconsideration and prompt allowance of claims 1-10 are earnestly solicited.

Should the Examiner believe that anything further would be desirable in order to place this application in even better condition for allowance, the Examiner is invited to contact the undersigned at the telephone number set forth below.

Respectfully submitted,



William P. Berridge
Registration No. 30,024

Jeremy D. Tillman
Registration No. 62,639

WPB:JDT/hs

Attachment:

Rule 1.132 Declaration with Polimery Publication

Date: April 15, 2009

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PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of

Inventor:	Johannes Bos	Group Art Unit:	1796
Application No.:	10/580,018	Examiner:	G. LISTVOYB
Filed:	May 19, 2006	Docket No.:	127898
For:	PROCESS FOR MAKING DAPBI-CONTAINING ARAMID CRUMBS		

DECLARATION UNDER 37 C.F.R. §1.132

I, Johannes Bos, a citizen of The Netherlands, hereby declare and state:

1. I have a degree as engineer in HTS-Chemistry, which was conferred upon me by Van't Hoff Instituut in Rotterdam, The Netherlands in 1984.
2. I have been employed by Teijin Aramid (formerly named Akzo, Akzo Nobel and Teijin Twaron), the Netherlands, since 1989 and I have had a total of 19 years of work and research experience in the field of aramid polymers for use in fiber spinning processes, as well as a range of other topics related to polymer science and engineering.
3. I am a named inventor in the following patents in this field: U.S. Patent No. 6,355,094; U.S. Patent No 5,776,354; U.S. Patent No 5,750,030; U.S. Patent No 5,738,791; WO 2008/028605 and WO 2008/061668.
4. I am a named inventor in the above-captioned patent application. I am familiar with the patent application.
5. I am employed by the Assignee, Teijin Aramid, of the above-identified

patent application. In the course of my employment, I receive compensation for my work relating to research and development regarding aramid polymers for use in fiber spinning processes. I am not being specially compensated for my work in preparing this Declaration.

6. The following experiments were included in internal records of Teijin Aramid. I have read and understood write ups of the experiments in these records and below is my opinion regarding these experiments.

I. In Making A Crumb As Recited In Present Claims 1 And 10. The Use Of Lithium Chloride And Calcium Chloride Is Not Equivalent

In the Office Action, I understand the U.S. Patent Office to allege that Chernykh, RU 2,017,866, indicates the use of lithium chloride and calcium chloride to be equivalent. See Office Action, page 3. Chernykh mentions the use of "CaCl₂ or LiCl" without any reference to make clear what the "or" actually means. The use of the word "or" does not necessarily mean that when CaCl₂ is changed for LiCl, all properties and effects remain the same. For instance, the viscosity of the polymer solution, relative viscosity of the polymer, flow behavior of the polymer solution etc. can be different. See the experimental results further in this declaration. Thus "or" does not mean that both salts are exchangeable without changing at least some of the results and effects.

In addition Chernykh relates to polymer solutions (page 4, column 2). Addition of CaCl₂ or LiCl both enhances the dissolving power. The exchangeability of CaCl₂ and LiCl as disclosed by Chernykh only relates to conditions to obtain polycondensate solutions that are suitable for extrusion and fiber spinning (page 4, col. 2).

However, lithium chloride and calcium chloride behave differently when making an

aramid crumb, and cannot be interchanged successfully as alleged by the examiner.

Four sets of polymerization experiments were conducted to show that calcium chloride and lithium chloride are not equivalent as compounds for forming aramid crumb.

A. First Set Of Experiments

The first set of experiments consisted of reproducing the traditional approach of forming a standard aramid poly-para-phenyleneterephthalamide (PPTA) crumb using p-phenylenediamine (PPD) and terephthaloyl dichloride (TDC) as the monomers. These experiments included taking ground and dried calcium chloride and stirring it into a reaction vessel (Waring Blender with a cooled bottom) containing N-methyl pyrrolidone (NMP) as the solvent.

The mixture of calcium chloride and NMP was continuously stirred during thermostating to 0 °C in order to obtain a finely divided suspension. Subsequently, PPD, distilled in vacuo (b.p. 168 °C/18 mm) and ground, was added and the mixture was continuously stirred. Shortly afterwards, molten TDC (distilled in vacuo, b.p. 136 °C/10 mm) was rapidly added from a dropping funnel to form a reaction mixture. The reaction mixture was heated with an IR lamp and vigorously stirred to prevent solidification. After about a minute, the reaction mixture formed a semi-solid material that separated into crumbs when the inherent viscosity (η_{inh}) of the polymer exceeded a value of about 3.

Table 1 illustrates the reaction conditions of the above experiment using different weight percentages of calcium chloride and the monomers (i.e., PPD and TDC). The resulting crumbs were coagulated with water, washed and then dried.

Table 1: Inherent Viscosity of First Set of Experimental Trials

Experiment No.	ml NMP	CaCl ₂ , wt% ¹	PPD and TDC monomer concentration, wt% ²	η_{inh}
1	200	15.25	11.84	4.05
2	200	5.66	9.09	3.05
3	200	7.41	8.94	3.15
4	200	9.09	8.79	4.05
5	200	10.71	8.65	4.02
6	200	12.28	8.51	5.05
7	200	16.67	8.12	4.6
8	200	20.00	7.82	3.8
9	40000	10.91	7.94	5.33

¹ $\text{wt}(\text{CaCl}_2)/(\text{wt}(\text{CaCl}_2)+\text{wt}(\text{NMP}))*100$.

² $\text{wt}(\text{monomers})/\text{wt}(\text{total mixture})*100$.

^{*} performed in a 160 L reactor.

As shown above in Table 1, a PPTA crumb was formed from a relatively low monomer concentration in the NMP/calcium chloride mixture and the inherent viscosity (η_{inh}) of the PPTA polymer was always greater than 3.

B. Second Set Of Experiments

A second set of experiments was performed in exactly the same manner as the first set of polymerization experiments, except that the CaCl₂ was replaced with CaBr₂, BaCl₂, CaSO₄, MgCl₂, ZnCl₂, Zn(OAc)₂, NH₄Cl, or LiCl, in the range of 3 – 15 wt%. The results for this second set of experimental trials are summarized below in Table 2.

Table 2: Inherent Viscosity of Second Set of Experimental Trials

Salt	Salt Concentration %	Monomer Concentration %	η_{inh}
CaBr ₂	10.71	8.65	0.73
CaSO ₄	10.71	8.65	0.39
MgCl ₂	2.91	9.33	0.44
MgCl ₂	5.66	9.09	0.56
MgCl ₂	8.26	8.86	1.39
MgCl ₂	10.71	8.65	0.95
MgCl ₂	13.04	8.44	0.44
ZnCl ₂	2.91	9.33	0.48
ZnCl ₂	5.66	9.09	0.58
ZnCl ₂	8.26	8.86	0.64
ZnCl ₂	10.71	8.65	0.59
ZnCl ₂	13.04	8.44	0.39
NH ₄ Cl	10.71	8.65	0.28
LiCl	10.71	8.65	0.30
BaCl ₂	10.71	8.65	0.29
Zn(OAc) ₂	9.91	8.72	0.19

As shown above in Table 2, the η_{inh} for a low monomer concentration using salts other than calcium chloride ranged from 0.19 to 1.39, which as described above, is too low of an inherent viscosity to form a crumb. Furthermore, the addition of LiCl only resulted in an inherent viscosity of 0.30, whereas as shown above, the addition of calcium chloride unexpectedly achieved a substantially greater inherent viscosity that enabled crumb formation.

C. Third Set Of Experiments

The third set of experiments consisted of a large number of polymerization reactions using a relatively high monomer concentration of PPD and TDC and using CaCl₂, LiCl, LiBr, MgCl₂ and MgBr₂ as the salts.

More specifically, the procedure for this third set of experimental trials consisted of charging an oven-dried reactor to a temperature of 60 °C, wherein the reactor contained:

450 mL (461.7 g) of NMP and 14.80 g of salt. The reactor was then flushed with nitrogen gas for 15 minutes to dissolve the salt under stirring. After cooling the reactor to 20 °C, 37 grams of PPD were added followed by the addition of 69.44 grams of TDC.

The TDC was either

(1) added to the mixture at once or (2) added in ten equal portions of 6.944 g.

If ten equal portions of the TDC were added to the mixture, the time between the addition of each portion of TDC was about 3 to 5 minutes and the temperature of the mixture increased after the addition of each portion of TDC.

After all portions of the TDC were added to the mixture, the mixture was stirred for 1 hour and 16.35 g of the indicated salt as powder were added to form the resulting mixture that coagulated into a slurry upon the addition of water. The slurry was filtrated over a glass filter and the polymer was dried in vacuo for 16 hours at 110 °C. The η_{inh} for each of the polymers was determined, and the results are summarized below in Table 3.

Table 3: Inherent Viscosity of Third Set of Experimental Trials

Exper. No.	PPD (grams)	TDC (grams)	Number of portions of TDC Added	Monomer Conc. (wt%)	Salt	Salt Conc. (wt.%)	End temp °C	Solvent	η_{inh}
1	37.00	69.44	1x	18.26	CaCl ₂	3.11	-	NMP	0.57
2	37.00	69.44	10x	18.26	CaCl ₂	3.11	74	NMP	0.43
3	37.00	69.44	10x	18.26	CaCl ₂	3.11	58	NMP	0.46
4	37.00	69.44	1x	18.26	CaCl ₂	3.11	56	NMP	0.63
5	37.00	69.44	10x	18.26	LiBr	3.11	58	NMP	0.38
6	37.00	69.44	10x	18.26	LiCl	3.11	60	NMP	0.57
7	25.82	48.15	5x	13.51	LiCl	2.19	60	NMP	0.41
8	37.00	69.44	10x	18.26	MgCl ₂	3.11	56	NMP	0.31
9	37.00	69.44	10x	18.26	MgBr ₂	3.11	60	NMP	0.30

As shown above in Table 3, each of the experimental trials, comprised of PPD, TDC, NMP and one of the various salts (including CaCl₂), had an η_{inh} value between 0.38 and 0.63. As the final products for this third set of experimental trials formed a turbid yellow to a brown syrupy mass, a crumb could not be formed from any of these experiments.

With regards to CaCl₂, the major difference between these experiments and the experiments of Table 1 is the monomer and the salt concentrations. In this case the amount of salt was too low and the amount of monomer too high to keep the polymerization going on.

Various LiCl concentrations and monomer concentrations were used as well, however, in none of the cases crumb was formed and no polymer with sufficiently high molecular weight was obtained.

D. Fourth Set Of Experiments

The fourth set of experiments repeated the same conditions as the first and second set of experiments. Here, the LiCl and CaCl₂ salt were added to the TDC/PPD mixture with either dimethyl acetamide (DMAc) or NMP as solvent. The η_{inh} for the fourth set of experiments is shown below in Table 4.

Table 4: Inherent Viscosity of Fourth Set of Experimental Trials

Solvent	Salt	Salt wt%	Monomer Concentration (weight %)	Appearance of Mixture	η_{inh}
DMAc	LiCl	0.50	9.54	Custard-like	0.89
		0.99	9.50	Custard-like	0.90
		2.91	9.33	Custard-like	0.78
		5.66	9.09	Custard-like	0.32
NMP	LiCl	0.50	9.54	Custard-like	0.28
		0.99	9.50	Custard-like	0.69
		2.91	9.33	Custard-like	0.74
		5.66	9.09	Custard-like	0.26
DMAc	CaCl ₂	0.99	9.50	Custard-like	0.24
		1.96	9.41	Custard-like	0.50
		2.91	9.33	Custard-like	1.05
		5.66	9.09	Custard-like	1.52
		8.26	8.86	Custard-like	1.56
		10.71	8.65	Custard-like	1.36
NMP	CaCl ₂	10.7	8.65	crumb	4.27*
					4.04
					4.31

*: Analysis performed in triplicate

As shown above in Table 4, the NMP/CaCl₂ solvent/salt combination had an η_{inh} value of 4.31, while the remaining solvent/salt combinations, including NMP/LiCl, only gave low molecular weight materials wherein the highest η_{inh} value was 1.56 (for DMAc/CaCl₂). As such, only the NMP/CaCl₂ solvent/salt combination obtained η_{inh} value

which was high enough to form a crumb.

The publication of E. Chodkowski (Polimeri, 1971, 514-515; English translation added as Annex) mentions the use of LiCl and CaCl₂ for the preparation of PPTA in DMAc. In case the salt concentration is between 0.5 – 1 mole/L the reduced viscosity (η_r) of PPTA with CaCl₂ is always significantly higher than η_r obtained with LiCl. Crumb formation is not disclosed (Table 5).

Table 5

Salt	Conc. salt, mole/L	Reduced viscosity
LiCl	0.5	0.808
LiCl	0.75	0.555
LiCl	1.00	0.372
CaCl ₂	0.50	1.539
CaCl ₂	0.75	1.273
CaCl ₂	1.00	0.497

E. Conclusions

The above results show that the formation of PPTA from TDC and PPD using CaCl₂/NMP solutions renders sufficient inherent viscosity (i.e. sufficient molecular weight). In case of LiCl the inherent viscosity is significant lower. As such, the above evidence confirms that the use of a CaCl₂/NMP salt/solvent combination for forming an aramid crumb is the exception. The rule is that no crumb is formed, as has been demonstrated by the use of other salt/solvent combinations, such as the LiCl/NMP salt/solvent combination.

F. Fifth Set Of Experiments

The fifth set of experiments relates to DABPI-containing aramids. With regard to aramids based on PPD, DAPBI and TDC, the situation is even more extreme. No documents were found that disclose the formation of crumbs with these DABPI-containing aramids. All published documents disclose that the polymer was obtained as a solution. These documents also suggested adding solubility promoting salts i.e. CaCl_2 or LiCl , to improve the solubility of the aramid in the solvent. See further also III.

In Table 6 below some comparisons are made between experiments 1 and 2 as performed by the R&D of Teijin Aramid and reference data from the prior art showing the results in different solvent systems and of different copolymer compositions.

Table 6

Experiment	Amines	Composition %	Solvent	Salt	b.c.	Appearance
1	PPD/DAPBI	50/50	NMP	CaCl_2	400	solution
2	PPD/DAPBI	50/50	NMP	LiCl	150	solution

Reference	Amines	Composition %	Solvent	Salt	b.c.	Appearance
Chernykh; example 1	CI-PPD/DAPBI	40/60	DMAc	LiCl	179	solution
Chernykh example 4	CI-PPD/DAPBI	80/20	DMAc	LiCl	62	solution
Chernykh example 7	PPD/DAPBI	70/30	DMAc	none	0	solution
Nakagawa US 4,018,735 example 4	PPD/DAPBI	90/10	NMP/HMP ¹	LiCl	50	semi-transparent
Yung example 8	PPD/DAPBI	40/60	NMP	none	0	solution
Mera US 4,172 938 example 34	PPD/DAPBI	80/20	NMP	CaCl_2	39	powder (see Table 8)

¹ hexamethylenephosphoramidate

In view of the fact that those of ordinary skilled in the art have not succeeded in forming PPTA crumbs using NMP/LiCl instead of NMP/CaCl₂, it is understood that NMP/LiCl will not give crumbs in DAPBI-containing aramid polymers. This has been confirmed by Example 4 of U.S. Patent No. 4,018,735 (Nakagawa), detailing the formation of a semi-transparent aramid dope from the reaction of PPD, DAPBI, TDC and a NMP/LiCl solvent and by Example 34 of U.S. Patent No. 4,172,938 (Mera) disclosing the formation of a powder (see Table 8).

7. I have conducted the following experiments.

II. Crumbs Are Obtained When Using The Conditions Recited In Present Claims 1 And 10

As described above, the copolymerization of PPD and TDC leads to an aromatic polyamide (PPTA) in the form of a crumb. Such crumbs are known and highly desired because they can be easily purified and possess a high processability for an extruder in a large scale commercial process. However, the present application embodies the scenario when a portion of PPD has been replaced with 5(6)-amino-2-(p-aminophenyl)-benzimidazole ("DAPBI"). Such compounds were found to form a solution, powder or gel, which has been a serious problem for large-scale production of a DAPBI-containing aramid.

To demonstrate the difficulty in obtaining a crumb from a DAPBI-containing polymer, I have performed two additional sets of experiments to show that crumbs are formed if the components and conditions recited in present claims 1 and 10 are satisfied.

The sixth set of experiments is represented as Examples 1-7 as described on page 5 of the present specification and the seventh set of experiments is represented as Comparative Examples 1-5 also described on page 5 of the present specification.

A. Sixth Set Of Experiments

The sixth set of experiments included pre-drying a 2L flask for 1 hour in an air circulation oven at 120 °C. Subsequently, the 2L flask was connected with a mechanical stirrer, an N₂ inlet and outlet stream between about 40 - 60 mL/min, and a vacuum supply. 400 mL of the NMP solvent containing 10.4 weight percent of CaCl₂ was combined with an amine mixture comprised of 90 mole percent PPD and 10 mole percent DAPBI and placed in a reactor. The flask was sealed and purged two times with nitrogen gas. After the amine mixture/NMP solvent were stirred for 30 minutes at 150 rpm, heated to 60 °C and mixed for 0.5 hours to dissolve or disperse the amines into the NMP solvent, the flask was cooled with a coolant to 5-10 °C. After removing the coolant, the stirrer velocity was increased to 320 rpm and 100 mole percent of TDC were introduced into the flask with a funnel. After the flask and the funnel were rinsed with 50 mL of the NMP solvent, the flask was closed and the mixture was allowed to react for at least 30 minutes. The whole reaction mixture spontaneously turned into a crumb. Such "crumb-particle" comprises polymer, solvent, salt, HCl and remaining monomers and oligomers. During reaction the monomer and oligomer concentrations diminish, so that when the process is stopped a crumb particle consists essentially of NMP, CaCl₂, HCl and polymer.

This aramid crumb was combined with demi-water in a Condux LV15 coagulator allowing the mixture to coagulate and was then collect on a RVS filter. The coagulated

crumb product was then washed four times with 5L of demi-water to remove the solvent, CaCl_2 and HCl and the washed product was collected in a 2L glass beaker and dried in vacuum for 24 hours at 80 °C.

Finally, the coagulated crumb was dissolved in sulfuric acid to form a solution to determine the relative viscosity and the inherent viscosity of the aramid crumb/sulfuric acid solution at 25 °C in an Ubbelohde viscometer. The above experiment was repeated six times with different mole percents of PPD and CaCl_2 . The results of each of these seven experiments are summarized below in Table 7.

Table 7: Process Conditions for Experiments 1-7

Experiment	PPD (a) mole%	DAPBI (b) mole %	CaCl_2 (c) wt%	b.c	η_{rel}	η_{inh}	Appearance
1	90	10	10.40	104.0	6.29	6.46	crumb
2	90	10	11.55	115.5	5.93	6.2*	crumb
3	80	20	9.85	197.0	5.38	5.92	crumb
4	80	20	10.28	205.6	4.10	5	crumb
5	60	40	4.77	190.8	5.69	6.01	crumb
6	33	67	3.09	207.0	6.98	6.45	crumb
7	30	70	2.82	197.4	6.2*	6.3*	crumb

average of 3 values

* calculated value

As shown above in Table 7, Experiments 1-7 each contain amounts of DAPBI ranging from 10 to 70 mole% and amounts of calcium chloride ranging from 2.82 to 11.55 wt%. Furthermore, each of the compositions described in Experiments 1-7 formed a crumb.

B. Seventh Set Of Experiments

The seventh set of experiments consisted of five comparative experiments. Comparative Experiments I-IV were performed under the same conditions as Experiments 1-7 in the sixth set of experiments, except that the amounts of DAPBI and calcium chloride were changed. Comparative Experiment V was performed in the same manner as described in U.S. Patent No. 4,172,938. Below Table 8 illustrates the results of the second set of experiments.

Table 8: Process Conditions for Comparative Experiments I-V

Comparative Experiments	PPD (a) mole%	DAPBI (b) mole %	CaCl ₂ (c) wt. %	b.c	η_{rel}	η_{inh}	Appearance
I	80	20	11.55	231.0	4.59	5.3	dough/paste
II	60	40	5.49	219.6	5.87	6.2	dough/paste
III	33	67	4.56	306.9	2.75	3.58	dough/paste
IV	33	67	2.88	193.0	2.31	3.04	gel
V	80	20	1.96	39.2	1.56	1.93	powder

The difference of comparative experiment IV of Table 8 and experiment 6 of Table 7 is the formation of a gel rather than a crumb, which is quite sensitive on the CaCl₂ concentration. If a gel is formed, then there is no further mixing and as a result movement of the reactive ends of the polymer to each other is inhibited whereby the reaction rate considerably drops thereby resulting in low molecular weight material.

As shown above in Table 8, when the mole percent of DAPBI (b) changes within the range from 20 to 67 mole% and the weight percent of calcium chloride (b) within the range from 1.96 to 11.55 wt%, no crumb is formed but a dough/paste, gel or powder, which are unsuitable for forming aramid fibers and films on a commercial scale. These data demonstrate that although the individual ranges of DAPBI and calcium chloride as

shown in Table 8 are similar to the ranges of Table 7, the relation between these values as expressed by the mole percent of DAPBI (b) multiplied by the weight percent of calcium chloride (c) is essential for the question whether or not a crumb will be formed.

C. Conclusion

It can therefore be concluded that replacing a portion of the PPD monomer with DAPBI in NMP/CaCl₂ under reaction conditions that are different from the conditions recited in claims 1 and 10 of the above-identified application as indicated in Table 8, will not form a crumb. In other words, crumbs can be obtained using this system, but a very specific relationship between the amounts of DAPBI and of CaCl₂ is required, which conditions are recited in the claimed subject matter.

III. Deficiencies Of Cited References

8. I have further read and understood the three references U.S. Patent No. 4,308,374 ("Vollbracht"), RU 2,017,866 ("Chernykh ") (English translation) and U.S. Patent No. 5,646,234 ("Jung") cited by the U.S. Patent Office in the Office Action. Below are my comments regarding each of these references.

Vollbracht, Jung and Chernykh teach away from the subject matter described in present claims 1 and 10. Vollbracht describes a method of producing PPTA using PPD and TDC in a NMP/CaCl₂ solvent/salt combination, wherein the method directly polymerizes PPD and TDC, turning the reaction mixture into a crumb during the polymerization reaction for various process conditions, as confirmed by the first set of experiments described above.

Jung teaches the addition of alkali metal and alkali earth metal halides, such as

lithium chloride and calcium chloride, as solubility-promoting additives (col. 3, lines 24-35). Jung adds these salts to keep the polymer in solution if the solubility of the polymer is too low to keep the polymer in solution without the addition of such a salt, even though such addition is not supported by experimental evidence. In Example 8 of Jung, DABI (= DAPBI) is used in a polymerization reaction performed according to the method of Example 1, i.e., without using a solubility-promoting additive. Thus, the solubility of the polymer described in Example 8 is high enough to not require the addition of a solubility-promoting additive. If nevertheless such a solubility-promoting additive is added, it would have been expected, in view of Jung's statements, that the solubility of the polymer of Example 8 would have been enhanced further.

Chernykh is similar to Jung in this respect. In Example 1 of Chernykh, 2.8% LiCl were added. This leads to a polymer solution, which can be filtered, degassed and spun to a fiber. As such, in Chernykh, LiCl has therefore been added as a solubility-promoting additive, and thus would not have been expected to lead to precipitation of the polymer.


Furthermore, the Patent Office alleges that Table 2 of Chernykh describes the combination of NMP and CaCl_2 . However, Table 2 of Chernykh clearly refers to Example 5 of Chernykh, which describes a benzothiazole derivative (5-amino-2-p-aminophenylbenzothiazole (DABPT)) rather than the benzimidazole derivative (i.e., DABPI), as recited in present claims 1 and 10. As the present claims are limited to the DABPI-containing aramids produced from DABPI monomeric building blocks, making a prediction that the solvent/salt combination (NMP/ CaCl_2) for the DABPT-containing aramids of Example 5 would be applicable to DABPI-containing aramids is clearly unsupported by the disclosure of Chernykh.

According to the process of present claims 1 and 10, the polymer should not remain dissolved in the solvent nor precipitate from the solvent. The reaction mass should follow the same reaction path as for PPD and TDC to form PPTA, i.e. after dosing and dissolving TDC, the viscosity of the reaction mass must increase during the reaction, the flowable mixture spontaneously turns into a dough or "liquid cheese-like" material, which spontaneously becomes a crumb. Thus, if an ordinarily skilled person in the art had the objective to provide a system wherein the polymer had to be obtained as a crumb (similar to Vollbracht), he or she certainly would not have added an alkali metal or alkali earth metal salt, because Jung and Chernykh describes that such addition increases the solubility of the polymer and thus would not have been expected to produce an crumb, which is not a solution.

In view of Jung and Chernykh, it was remarkable and unexpected that the use of calcium chloride in DABPI based polymer mixtures according to the present claims does not enhance the solubility of the polymer, but to the contrary leads to the formation of an aramid crumb during copolymerization.

9. I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Date: 24 - 3 - 2009



Johannes Bos

Translation of an article in Polimery, 1971, pp. 514-515 of Edward Chodkowski, Jan Mackowiak, Wojciech Kozlowski, Hanna Orzechowska of Instytut Włókien Sztucznych i Syntetycznych (Institute for Artificial and Synthetic Fibres). Lodz, Pol.

Solution polycondensation of terephthaloyl chloride and m- and p- phenylene diamine in the presence of LiCl, LiBr and CaCl_2 .

Aromatic polyamides may be obtained by interfacial polycondensation (1) of aromatic chlorides derived from a dicarboxylic acid and aromatic diamines, or by emulsion polycondensation (2) or by polycondensation in a homogeneous medium of polar organic solvents (3).

Of the greatest interest is to be considered the polycondensation in a medium of amide solvents such as dimethyl formamide, dimethyl acetamide or N-methyl pyrrolidone, which also act as acceptors of the hydrogen chloride evolved in the reaction. In the solution polycondensation addition of an appropriate amount of some salt selected from the potassium or the calcium group to some solvents (4) makes it possible to prepare solutions of aromatic polyamides in a concentration of as high as 15-20%.

The aromatic polyamide solutions thus obtained can be used in the manufacture of films or fibres having a very high thermal resistance (5-6). Our investigations comprise the polycondensation of terephthaloyl chloride and m- and p-phenylene diamine in a medium of dimethyl acetamide at different concentrations of lithium chloride, lithium bromide or calcium chloride. In the preparation of the respective aromatic polyamides (6) considerable attention has already been paid to the use of lithium salt and other alkali metal salts, but the use for this purpose of calcium chloride is only mentioned in a few patents (4).

Experiments.

According to Coppinger (7) N,N-dimethyl acetamide was obtained from acetic anhydride and dimethyl formamide in a yield of 62% of the theoretical value. Boiling temperature 165°C at 760 mm Hg.

Terephthaloyl chloride was obtained from thionyl chloride and commercial terephthalic acid (8) in 75% yield, melting point $77^{\circ}\text{--}78^{\circ}\text{C}$ (recrystallization from petroleum ether).

Commercial m-phenylene diamine was purified by sublimation in an atmosphere of nitrogen under reduced pressure (10 mm Hg). The main fraction sublimates at a temperature of $147^{\circ}\text{--}148^{\circ}\text{C}$. Melting point 63°C .

Commercial p-phenylenediamine was purified by sublimation in an atmosphere of nitrogen. The white acicular crystals are formed at a temperature of $140^{\circ}\text{--}142^{\circ}\text{C}$. Beforehand lithium chloride, lithium bromide and calcium chloride had been heated to 400°C for 2 hours.

Polycondensation was carried out in a 100-ml conical flask provided with a stirrer, thermometer, nitrogen inlet tube and a reflux condenser and an outlet tube filled with calcium chloride. After removal of the air with the aid of dry nitrogen the flask was charged with 25 ml of dimethyl acetamide and a measured quantity of LiCl , LiBr or CaCl_2 , followed by stirring to obtain a homogeneous solution in which 1.458 g (0.0135 moles) of m- or p-phenylene diamine were dissolved under the same conditions. The flask was cooled to a temperature of -8° to -10°C , and over a period of 20 minutes there were gradually added, with vigorous stirring, 2.745 g (0.0135 moles) of terephthaloyl chloride, after which stirring was continued for 40 minutes. The polymer was isolated from the mixture by pouring the latter into water or methanol (500 ml), with constant stirring, followed by sucking it off through a glass Büchner funnel, washing it with methanol or acetone (in the case where it is poured into water) and subsequently drying it to a constant weight at a temperature of 110°C .

The reduced viscosity of the resulting polymers was determined from viscosity measurements, using an Ubbelohde viscometer at a temperature of 25°C, and polymer solutions in 95% sulphuric acid in a concentration of 0.5 g/100 ml of acid.

The thermal resistance of the polyamides was determined with the aid of a dynamic differential calorimeter (Perkin-Elmer DSC-1 B) and their infrared spectrum, in the range of 4000-650 cm^{-1} , was determined with the aid of a spectrophotometer (Highler & Watts Model H-800), use being made of KBr tablets.

Discussion of the results.

From the results of initial experiments in which polyphenylene terephthalamide was isolated from the reaction mixtures with the aid of water or methanol it appears that for this purpose it is far more economical to use distilled water. Polymers obtained by isolation with the aid of methanol are whiter, but the colour of the polymers isolated by means of water can be improved by rinsing the moist precipitate in acetone and subsequently drying it in an atmosphere of nitrogen, which is consequently effected very fast.

Table I.

The yield of polyphenyleneterephthalamide as a function of the precipitation medium (solution polycondensation in dimethyl acetamide without the addition of a lithium salt or calcium salt).

Name of polymer	precipitation medium	polymer yield	reduced viscosity
poly-m-phenylene-terephthalamide	methanol	65.8	0.299
		67.1	0.397
		68.2	0.300
		71.1	0.240
poly-p-phenylene-terephthalamide	water	39.3	0.480
		90.0	0.300
		91.3	0.440
		92.7	0.300

Isolating and washing polyphenyleneterephthalamide with methanol does not have any positive influence on the viscosity of this polymer, which might be expected if the low-polymeric fraction of oligomers should be soluble in this solvent (Table I)

The yield and the molecular weight of aromatic polyamides obtained by solution polycondensation are clearly influenced by the addition to the reaction medium of particular salts from the potassium group, more particularly the chloride and the nitrate of lithium (5,6).

Our investigations show that the use for this purpose of calcium chloride may lead to results similar to those obtained in the case where lithium salts are employed. The average yield of polyphenyleneterephthalamide both in the presence of a lithium salt and in the presence of calcium chloride (isolated in water and washed with acetone) is 95-98% of the theoretical value, no difference having been found between the isomeric aromatic diamines, the viscosity of the polyamides, however, is to a certain extent dependent on the nature of the isomer. The thermal characteristic of the samples of poly-m-phenyleneterephthalamide and poly-p-phenyleneterephthalamide (Table II, runs 17 and 20)

is represented in a thermogram (Fig.1) obtained with the use of a dynamic calorimeter (DSC).

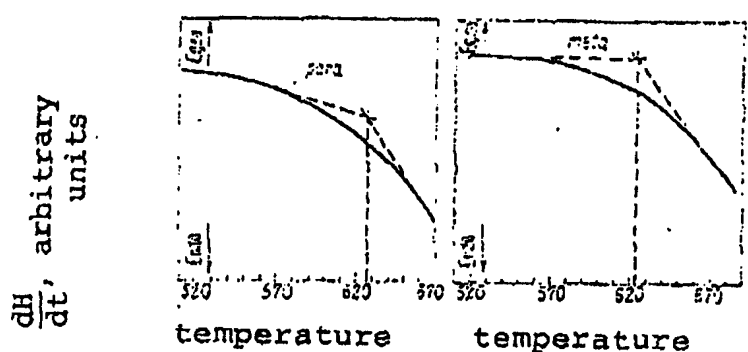


Fig.1. DSC thermograms for poly-m-phenylene terephthalamide (right) and poly-p-phenylene terephthalamide (left).

The thermograms show no sharp changes as regards the development (crystallization) or absorption (melting) of heat, which is characteristic of crystalline polymers. The smooth inflection of the thermogram curve into the endothermal direction only points to a slow thermal decomposition of these polymers, which is distinctly speeded up at a temperature $>350^{\circ}\text{C}$. A similar characteristic is found by visual observation. For instance during the slow heating process (about $2^{\circ}\text{C}/\text{minute}$) in a capillary the meta-isomer turned yellow at a temperature of 325°C and it softened at a temperature of about 352°C , clearly displaying decomposition. The poly-m-phenyleneterephthalamides have a lower viscosity than the poly-p-phenyleneterephthalamides obtained under the same conditions in the presence of a lithium salt or a calcium salt (Table II).

Table II synthesis of poly-m- and p-phenyleneterephthalamides.

Name of diamine	Salt type and concentration		Reduced viscosity	
		concentration		η_r
		mole/l	%	
m-phenylenediamine	-			0.240
p-phenylenediamine	-			0.300
m-phenylenediamine	-			0.440
p-phenylenediamine	-			0.315
m-phenylenediamine	LiCl	0.50	2.12	0.460
	"	0.75	3.18	0.379
	"	1.00	4.24	0.338
p-phenylenediamine	"	0.50	2.12	0.750
	"	0.75	3.18	0.420
	"	1.00	4.24	0.338
m-phenylenediamine	LiBr	0.50	4.34	0.610
	"	0.75	6.51	0.473
	"	1.00	8.69	0.360
p-phenylenediamine	"	0.50	4.34	0.808
	"	0.75	6.51	0.555
	"	1.00	8.69	0.372
m-phenylenediamine	CaCl ₂	0.50	5.55	1.413
	"	0.75	8.33	0.718
	"	1.00	11.10	0.340
p-phenylenediamine	"	0.50	5.55	1.539
	"	0.75	8.33	1.273
	"	1.00	11.10	0.497

The infrared spectra of the isomeric aromatic polyamide samples are very much alike. Differences in absorption are only found in the range of 900-750 cm^{-1} .

Poly-m-phenyleneterephthalamide shows an intensive band in the vicinity of 850 cm^{-1} , poly-p-phenyleneterephthalamide, however, in the neighbourhood of 755 cm^{-1} , which is characteristic of the position of meta and para. Comparing the two spectra, we can establish that the peaks at the right of the spectrum of poly-p-phenyleneterephthalamide have shifted towards higher wave lengths than the isomeric m-polymer (Fig.2)

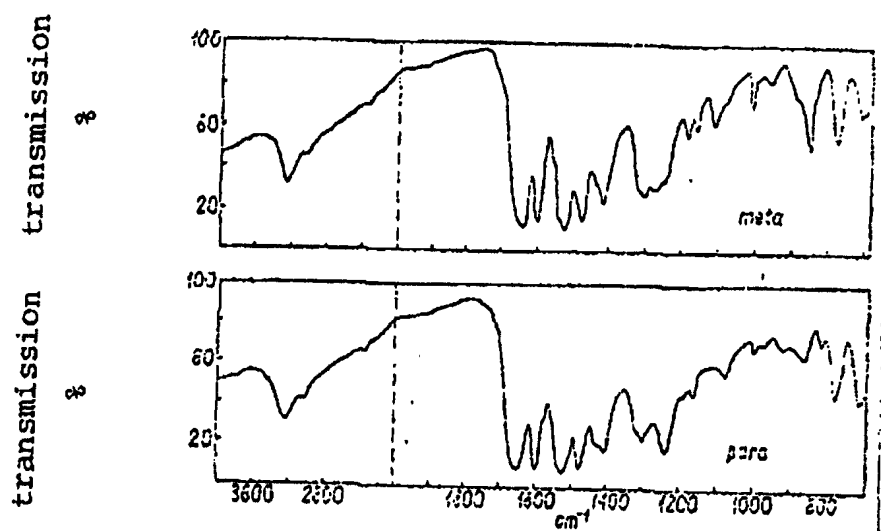


Fig. 2 Infrared spectrum of poly-m-phenyleneterephthalamide (top) and poly-p-phenyleneterephthalamide.

Acknowledgement is due to Prof. Dr. T. Skwarski for his interest in and discussion of the subject.

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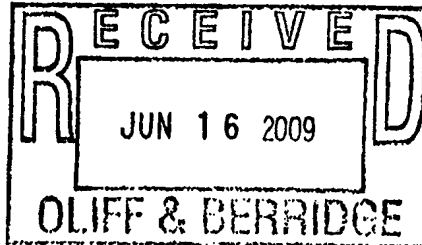


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Please find below and/or attached an Office communication concerning this application or proceeding.

The time period for reply, if any, is set in the attached communication.

**FINAL REJECTION
NOTICE OF APPEAL**

DUE DATE

SEP 15 2009

DOCKETED
By SCE on 6/16 2009
and
By BRJ on June 16 2009
Oliff & Berridge

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DETAILED ACTION

Claim Rejections - 35 USC § 103

The following is a quotation of 35 U.S.C. 103(a) which forms the basis for all obviousness rejections set forth in this Office action:

(a) A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negated by the manner in which the invention was made.

Claims 1-5 and 10 rejected under 35 U.S.C. 103(a) as being unpatentable over Vollbracht et al (US 4308374) herein in view of Vollbracht Chernykh et al (RU 2017866) herein Chernykh or Jung et al (US 5646234) herein Jung (both cited in the previous Office Action)

Vollbracht teaches a method for obtaining a an aromatic polyamide containing para-phenylene terephthalamide in the form of a crumb, by polymerizing para-phenylenediamine and terephthaloyl dichloride in a mixture of N-methyl pyrrolidone and containing calcium chloride

Vollbracht teaches is an aromatic polyamide in the crumb form, which has a relative viscosity of at least 4 and the crumb is formed directly by the copolymerizing (see Example 1).

Vollbracht teaches that his polyamide can be used for fiber formation.

Vollbracht does not teach 5(6)-amino-2-(p-aminophenyl)benzimidazole and ratios between the monomers, which are suitable for crumb formation.

Chernykh discloses a method for obtaining a composition for fiber formation comprising an aromatic polyamide containing para- phenylene terephthalamide and 2-(p-phenylene)benzimidazole terephthalamide units by copolymerizing: i) a= 10-80 mole % of para-phenylenediamine; ii) b=10-80 mole % of 5(6)-amino-2-(p-aminophenyl)benzimidazole; and iii) 100 mole% of terephthaloyl dichloride (see Example 1).

Note that in the Example 1 Chernykh uses Cl-para-phenylenediamine, whereas Claim 1 claims para-phenylenediamine. However, Chernykh teaches that the above compounds are equivalent (see Page 7, line 15).

Also, Chernykh teaches that DMAA and NMP as well as LiCl or CaCl₂ can be equally used (see Page 3, line 25).

In a mixture of N-methyl pyrrolidone and containing 2.8-4.8% wt.% of calcium chloride or lithium chloride (see Table 2, column 2), a + b is 100 mole% and i), ii), and iii) together comprise 1-20 wt.% of the mixture (see Example 1). Based on the above data the result of b x c multiplication can be less than 215.

Intrinsic viscosity of the polymer above is between 5.9 and 10.9 dl/g (see Table 2).

Jung discloses method of obtaining an aramid by polycondensation of 100 mol% terephthaloyl chloride, 40 mol% p-phenylenediamine and 60 mol% 5(6)-amino-2-(p-aminophenyl)-benzimidazole in N-methylpyrrolidone, wherein $a+b=100$ mol% and the relative viscosity is 4,3 (see example 8).

Jung discloses solubility-promoting additives such as calcium dichloride in amounts of between 0,2 and 10 wt.%, preferably between 0,5 and 5 wt.%, can be added to the polycondensation mixture (see column 3, lines 19-35 and 62-67).

Chernykh or Jung do not disclose a crumb obtained directly by copolymerization, since their processes are intended to directly process a solution into article immediately after synthesis.

5(6)-amino-2-(p-aminophenyl)benzimidazole is a common monomer for fiber-forming polymer. Typically this compound used when high temperature resistance or broad temperature range of application is needed. Also benzimidazole known for their fire resistance and good mechanical properties (especially tensile strength and modulus).

Relations between monomer content, CaCl_2 and other technological parameters in order to obtain a crumb can be adjusted by an artisan with routine experimentation approach.

Therefore, it would have been obvious to a person of ordinary skills in the art to apply 5(6)-amino-2-(p-aminophenyl)benzimidazole comonomer in Vollbracht's fiber-forming polyamide, since it increases temperature application range, enhances mechanical properties and possess excellent fire resistivity.

Claims 2-5 rejected under 35 U.S.C. 103(a) as being unpatentable over Chernykh et al (RU 2017866) herein Chernykh or Jung et al (US 5646234) herein Jung (both cited in the previous Office Action) in combination with Vollbracht et al (US 4308374) herein Vollbracht or Encyclopedia of Pol. Sci and Tech (Polyamides, vol 3, p. 565-567) herein Encyclopedia and the Applicant's admission of the Prior Art in the Specification ((both cited in the previous Office Action).

Chernykh discloses a method for obtaining a composition comprising an aromatic polyamide containing para- phenylene terephthalamide and 2-(p-phenylene)benzimidazole terephthalamide units by copolymerizing: i) a= 10-80 mole % of para-phenylenediamine; ii) b=10-80 mole % of 5(6)-amino-2-(p-aminophenyl)benzimidazole; and iii) 100 mole% of terephthaloyl dichloride (see Example 1).

Note that in the Example 1 Chernykh uses Cl-para-phenylenediamine, whereas Claim 1 claims para-phenylenediamine. However, Chernykh teaches that the above compounds are equivalent (see Page 7, line 15).

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Chernykh or Jung do not disclose a crumb obtained directly by copolymerization, since their processes are intended to directly process a solution into article immediately after synthesis.

Regarding Claims 4 and 5, Encyclopedia discloses a process of obtaining Poly(p- phenylene terephthalimide) in of N-methyl pyrrolidone at the presence of Calcium Chloride. Encyclopedia teaches a process for making a purified aromatic polyamide, which comprises coagulating and washing the crumb in water, followed by drying step (see pages 565 and 567).

Vollbracht teaches a method for obtaining a an aromatic polyamide containing para-phenylene terephthalamide in the form of a crumb, by polymerizing para-phenylenediamine and terephthaloyl dichloride in a mixture of N-methyl pyrrolidone and containing calcium chloride

Vollbracht teaches is an aromatic polyamide in the crumb form, which has a relative viscosity of at least 4 and the crumb is formed directly by the copolymerizing (see Example 1).

Vollbracht teaches that his polyamide can be used for fiber formation.

Chernykh or Jung does not teach the polymer precipitation step, since synthesis of polymer and its processing locates in one facility. However, more commonly those two processes are separated. In this case it is economically efficient to transport and store dried polymer instead of its diluted solution in N-methyl pyrrolidone.

Therefore, it would have been obvious to a person of ordinary skills in the art to precipitate Chernykh's or Jung's polymer using Encyclopedia's and Vollbracht's technology in order to expand applicability of the polyamide.

Chernykh or Jung and Encyclopedia does not disclose the crumb is defined as non-sticky particles at least 95% of which having an average diameter of 0.7-15 mm.

However, according to Applicant's discussion of the Prior Art in the Specification, such crumbs are known from the process of preparing of fully aromatic polyamides based on e.g. PPD and TDC, which products are known under the trade names Twaron® (Teijin Twaron) and Kevlar (DuPont). After polymerization in NMP/CaCl₂ a crumb is obtained which can be easily coagulated, washed, and dried, and the product obtained can be dissolved in sulfuric acid and shaped into a desired form, like fibers or films.

The crumbs of the above particles are very process-friendly, in particular, in a filtration step. The efficiency of the above step can be impaired with fines or sticky gels. Polymer precipitation aiming the crumbs with particular particle size can be achieved with well known technological approaches (rate of precipitant adding, temperature, stirring, etc).

Therefore, it would have been obvious to a person of ordinary skills in the art to obtain crumb with optimum particle size (i.e. 0.7-15 mm) in order to achieve efficient filtration process.

It is noted that there is no showing of unexpected results, associated with particular particle size range in the Specification.

Response to Arguments

Applicant's arguments filed 4/15/2009 have been fully considered but they are not persuasive.

Applicant argues that Vollbracht does not describe replacing a portion of the PPD with 5(6)-amino-2-(p- aminophenyl)-benzimidazole ("DAPBI") and it would not have been obvious to one of ordinary skill in the art to have combined the teachings of Chemykh with Jung to form the crumb obtained in Vollbracht using a different composition.

Examiner disagrees. It is clear that solubility of the polymer depends on its structure. Relations between monomer content, CaCl_2 and other technological parameters in order to obtain a crumb can be adjusted by an artisan with routine experimentation approach.

Regarding Chernykh, Applicant argues that "Chernykh Would Not Have Provided One With Any Reason or Rationale To Have Replaced A Portion of PPD With DAPBI And Form A Crumb".

However, the primary reference (Vollbracht) provides the teaching of forming the crumb during the polycondensation process.

In addition, Chernykh teaches that DARBI-based (benzimidazole containing polymers) have better tensile strength, Young modulus and other mechanical properties, compare to ones, based on regular aromatic polyamides (see page 2, lines 45-60).

Applicant argues that Chernykh's copolymer is not equal to one claimed. Applicant submits that in Example 1 the Reference uses LiCl instead of CaCl₂. In, in the Example polymer structure is different from one claimed.

However, Examiner relies on the Chernykh disclosure in its entirety, where both salts and claimed monomers are used.

According to MPEP 2123, disclosed examples and preferred embodiments do not constitute a teaching away from a broader disclosure or nonpreferred embodiments (see also *In re Susi*, 440 F.2d 442, 169 USPQ 423 (CCPA 1971), *In re Gurley*, 27 F.3d 551, 554, 31 USPQ2d 1130, 1132 (Fed. Cir. 1994) , *In re Fulton*, 391 F.3d 1195, 1201, 73 USPQ2d 1141, 1146 (Fed. Cir. 2004).

Applicant argues that "The Conditions For Obtaining A Crumb Directly During The Copolymerization Are Not Described In Chernykh." Applicant submits that the copolymerization of PPD and TDC leads to a PPTA crumb and that the

present application embodies the scenario when a portion of PPD has been replaced with DAPBI. Prior to the present application, such a replacement formed a powder compound or a gel compound, which has been a serious problem for large-scale production of a DAPBI- containing aramid.

Examiner disagrees. Firstly, in all cases (with or without PPD or DARBI) crumb formation strictly dictates by solubility of the polymer in a system. Therefore, crumb can be formed in both cases varying solvent/non-solvent concentration, depending on the polymer structure. In other words, polyamide, based on PPD can remain in the solution if the concentration of a precipitant is not sufficient.

Applicant argues that "The Patent Office's Allegation That Chernykh Describes The Equivalence Of LiCl And CaCl₂ Would Be Applicable To Crumb Formation Is Incorrect".

Applicant submits Declaration under 37 CFR 1.132, where required IV of the polymer is not achieved, when LiCl is used (see Table 2 of the Declaration). However, only two concentrations of the salts (i.e. 10.71 and 8.65%wt) was used for both LiCl and CaCl₂. This is not commensurate with the scope of the claims 1 and 2, which does not claim specific salt concentration.

Regarding Jung, applicant argues that Reference fails to describe the composition of claim 2, a composition in crumb form, or how to obtain the

composition of claim 2 in crumb form by the use of the process (solvents and amounts of materials) of claim 1 and claim 10.

Examiner disagrees. Jung discloses method of obtaining an aramid by polycondensation of 100 mol% terephthaloyl chloride, 40 mol% p-phenylenediamine and 60 mol% 5(6)-amino-2-(p-aminophenyl)-benzimidazole in N-methylpyrrolidone, wherein a+b=100 mol% and the relative viscosity is 4,3 (see example 8).

Jung discloses solubility-promoting additives such as calcium dichloride in amounts of between 0,2 and 10 wt.%, preferably between 0,5 and 5 wt.%, can be added to the polycondensation mixture (see column 3, lines 19-35 and 62-67).

Vollbracht (primary reference) teaches a method for obtaining an aromatic polyamide containing para-phenylene terephthalamide in the form of a crumb.

Conclusion

THIS ACTION IS MADE FINAL. Applicant is reminded of the extension of time policy as set forth in 37 CFR 1.136(a).

A shortened statutory period for reply to this final action is set to expire **THREE MONTHS** from the mailing date of this action. In the event a first reply is filed within **TWO MONTHS** of the mailing date of this final action and the advisory action is not mailed until after the end of the **THREE-MONTH** shortened statutory period, then the shortened statutory period will expire on the date the advisory

action is mailed, and any extension fee pursuant to 37 CFR 1.136(a) will be calculated from the mailing date of the advisory action. In no event, however, will the statutory period for reply expire later than SIX MONTHS from the mailing date of this final action.

Any inquiry concerning this communication or earlier communications from the examiner should be directed to GREGORY LISTVOYB whose telephone number is (571)272-6105. The examiner can normally be reached on 10am-7pm.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, James Seidleck can be reached on (571) 272-1078. The fax phone number for the organization where this application or proceeding is assigned is 571-273-8300.

Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see <http://pair-direct.uspto.gov>. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free). If you would like assistance from a USPTO Customer Service Representative or access to the automated information system, call 800-786-9199 (IN USA OR CANADA) or 571-272-1000.

Application/Control Number: 10/580,018

Art Unit: 1796

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GL

PROCESS FOR MAKING DAPBI-CONTAINING ARAMID CRUMBS

CROSS-REFERENCE TO RELATED APPLICATION

This application is a U.S. national stage application of PCT/EP2004/012760, filed November 11, 2004, which PCT application is incorporated herein by reference in its entirety.

BACKGROUND

The invention relates to a method for obtaining a composition comprising an aromatic polyamide containing para-phenylene terephthalamide and 2-(p-phenylene)-benzimidazole terephthalamide units by copolymerizing para-phenylenediamine (PPD), 5(6)-amino-2-(p-aminophenyl)benzimidazole (DAPBI); and terephthaloyl dichloride (TDC) in a mixture of N-methyl pyrrolidone (NMP) and calcium chloride, and to said composition. The invention further relates to a method for making purified aromatic polymer from said composition.

Methods for making aramid polymers are known in the art. For instance, in US 4,172,938, an aromatic polyamide was described to be made by polymerizing a mixture of diamines and an aromatic dicarboxylic acid dihalide in a mixture of N-methyl pyrrolidone and calcium chloride. In example 34 of this reference, the polymerization reaction is performed with a mixture of para-phenylene diamine (PPD) and 5-amino-2-(p-aminophenyl)-benzimidazole (DAPBI), and terephthaloyl dichloride (TDC) in N-methyl pyrrolidone (NMP) containing 2 wt.% of calcium chloride (CaCl_2). The product was obtained as a powdery clay-like material for which filtration was problematic. It was disclosed that products according to this reference in more general terms are obtained as slurry, paste, powder, or agar.

Other processes for making spin dopes of DAPBI-containing polymers are known from US 5,646,234 and US 4,018,735. US 5,646,234 discloses a process for making a spin dope wherein the use of alkali metal halides, among which also calcium chloride, as additive is disclosed. However, very particular preference is given in using no additives, and in conformity herewith the specific examples do not use such additives. Moreover, if calcium chloride is applied according to this reference the amount thereof can be substantially higher than allowable for preventing formation of paste and the like. None of the examples of US 4,018,735 disclose the use of calcium chloride, nor is such specific additive suggested in combination with N-methyl pyrrolidone, and for that reason the polymers of this reference will be obtained in the form of a paste, powder, and the like.

SUMMARY

It is an object of the present invention to provide conditions for performing such reaction and obtaining a composition in the form of a crumb or a crumb-like material. The term crumb or crumb-like as used in this invention means that the polymer mixture contains breakable clumps or particles, which are not sticky and have a mean particle size greater than 100 μm , usually greater than 1 mm. The term crumb in relation to this invention is defined as non-sticky particles, i.e., particles as in powders that do not stick together and remain free from each other, at least 95% of which have an average diameter 0.7-15 mm, preferably 1-7 mm.

DETAILED DESCRIPTION OF EMBODIMENTS

Such crumbs are known from the process of preparing of fully aromatic polyamides based on e.g., PPD and TDC, which products are known under the trade names TWARON® (Teijin Twaron) and KEVLAR® (DuPont). After polymerization in NMP/ CaCl_2 , a crumb is obtained which can be easily coagulated, washed, and dried, and the product obtained can be dissolved in sulfuric acid and shaped into a desired form, like fibers or films.

The monomer of interest, DAPBI (5(6)-amino-2-(p-aminophenyl)-benzimidazole; CAS reg. no: 7621-86-5), is added to the diamine mixture with the objective to obtain a suitable polymer solution right after polymerization with e.g., PPD and TDC, which can be directly shaped into fibers or films, whereby DAPBI is seen as a suitable co-monomer to keep the aramid polymer in solution. By selecting a specific ratio of PPD, DAPBI, and CaCl_2 , the formation of powders, paste, dough, and the like can be prevented.

To this end, the invention relates to a method for obtaining a composition comprising an aromatic polyamide containing para-phenylene terephthalamide and 2-(p-phenylene)benzimidazole terephthalamide units by copolymerizing:

- i) a mole % of para-phenylenediamine;
- ii) b mole % of 5(6)-amino-2-(p-aminophenyl)benzimidazole; and
- iii) 90-110 mole% of terephthaloyl dichloride

in a mixture of N-methyl pyrrolidone and containing c wt.% of calcium chloride, wherein c is within the range from 1 to 20, and wherein the ratio a : b ranges from 1 : 20 to 20 : 1, a + b is 100 mole%, and i), ii), and iii) together comprise 1-20 wt.% of the mixture, wherein the product b.c is at least 50 and less than 215 and that the composition is a crumb with a relative

viscosity η_{rel} of at least 4, wherein the crumb is defined as non-sticky particles at least 95% of which having an average diameter of 0.7-15 mm.

It is another objective of the present invention to obtain crumbs comprising a polymer with a sufficient high relative viscosity η_{rel} . Relative viscosities η_{rel} of at least 4, more preferably between 4 and 7, most preferably at least 5 can be obtained according to the method of the invention. It is further preferred to have a mixture for copolymerization wherein b.c is at least 80.

In another object of the invention a method for obtaining a purified aromatic polyamide is obtained by coagulating and washing the obtained crumb with water, followed by drying. The drying step can be performed according to standard procedures, such as ambient conditions, or at elevated temperature and/or lowered pressure. Thus, the obtained material is suitable for making a spin dope by dissolving it in a solvent, for instance sulfuric acid, NMP, NMP/ CaCl_2 , dimethylacetamide, and the like. The dope can be used to manufacture formed articles, such as fibers, films, and the like.

In the following experiments, the aspects of the invention are exemplified.

General polymerization procedure

DAPBI (ex Spektr T.T.T., Russia) was dried under vacuum for 1 h at 160° C. PPD (Teijin Twaron), TDC (freshly distilled), NMP/ CaCl_2 and NMP (both ex Teijin Twaron) were used as received (moisture content 80 ppm).

The glassware was pre-dried for 1 h in an air circulation oven at 120° C. A clean and dry 2 liter flask was supplied with a mechanical stirrer, N_2 -inlet and outlet, and vacuum supply. Generally, the N_2 -stream is between 40 – 60 ml/min. A large part (400 ml) of the solvent and the precisely pre-weighed amines were carefully brought in the reactor. The reactor was closed and purged two times with nitrogen. The mixture was stirred for 30 min at 150 rpm and heated to 60° C and mixed for 0.5 h to dissolve or disperse the amines properly. The flask was cooled with ice/water to 5 – 10° C. After removing the coolant, the stirrer velocity was set at 320 rpm and a precisely pre-weighed amount of the acid chloride was brought into the vessel through a funnel. In all cases the mol ratio of the total number of amines and the acid chloride equals one. The flask, which contained the acid chloride and the funnel, was rinsed with the remaining solvent (50 ml). The vessel was closed and the mixture was allowed to react for at least 30 min (nitrogen flush between 40 – 60 ml/min). The stirring was stopped and the reaction vessel was removed.

The crumbed product together with demi-water was gently added into a Condux LV15 15/N3 coagulator and the mixture was collected on an RVS filter. The product

was washed 4 times with 5 l of demi-water, collected in a 2 l glass beaker and dried under vacuum for 24 h at 80° C.

5 A sample was dissolved in sulfuric acid at room temperature. The flow time of the sample solution in sulfuric acid 96% (0.25 % m/V) was measured at 25° C in an Ubbelohde viscometer. Under identical conditions the flow time of the solvent was measured as well. The relative viscosity was then calculated as the ratio between the two observed flow times.

Results:

10 After addition of the TDC, the temperature increased rapidly and could reach its maximum between 40° and 70° C.

The Table shows some examples in which the polymer mixture turned into a crumbed mass, which could easily be coagulated and washed. To obtain crumbs the DAPBI content, monomer concentration, and the CaCl_2 concentration must be balanced according to the invention. The relative viscosity, inherent viscosity, and appearance (crumb or others) are
15 given in the Table. PPD, DAPBI and TDC together comprise about 10 to 12 wt.% of the mixture.

In comparative examples I – III, the polymer mixture was rendered as a dough polymer mass or as rubbery “chewing gum-like” mass, due to the high CaCl_2 content. In Comparative Example IV, first a precipitate was formed, which was later converted to a
20 dough-like mass. The CaCl_2 content was too low to obtain a crumb. Comparative Example V (according to US 4,172,938 having an inherent viscosity of 1.93, see Table) resulted in a powdery material, which after coagulation was very difficult to filter. It behaved like a clay-like material.

Table

Examples	PPD a mole%	DAPBI b mole %	CaCl ₂ c wt. %	b.c	η_{rel}	η_{inh}	crumb
1	90	10	10.40	104.0	6.29	6.46	yes
2	90	10	11.55	115.5	5.93	6.2 [#]	yes
3	80	20	9.85	197.0	5.38	5.92	yes
4	80	20	10.28	205.6	4.10	5	yes
5	60	40	4.77	190.8	5.69	6.01	yes
6	33	67	3.09	207.0	6.98	6.45	yes
7	30	70	2.82	197.4	6.2 [*]	6.3 [#]	yes
Comparative Examples							
I	80	20	11.55	231.0	4.59	5.3 [#]	dough/paste
II	60	40	5.49	219.6	5.87	6.2	dough/paste
III	33	67	4.56	306.9	2.75	3.58	dough/paste
IV	33	67	2.88	193.0	2.31	3.04	gel ⁺
V	80	20	1.96	39.2	1.56	1.93	powder

* average of 3 values

calculated value

+ gel with precipitated particles

The Table shows the advantageous properties when the conditions of the invention are satisfied. For instance, Comparative example V (according to US 4,172,938) has a product b.c value outside the claimed range (39.2), but a relative viscosity below 4 (1.56). No crumb is formed, but a powder is formed (having a particle size far below the average diameter 0.7 mm).

XIV. APPENDIX E - RELATED CASES APPENDIX

NONE